Materials Capability Review

Los Alamos National Laboratory

May 31 - June 3, 2011

On the cover: Phase-field simulation of gas bubble nucleation and evolution (helium, yellow) in a polycrystalline material (uranium oxide, blue) showing nearly continuous coverage of the grain boundaries plus scattered interior sites.

Contents

2011 Materials Capability Review Committee

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- W.R. Cieslak
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- A.J. Taylor

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Agenda

Materials Capability Review
Los Alamos National Laboratory

May 31-June 3, 2011

Hilton @ Buffalo Thunder 5th Floor Executive Lounge
30 Buffalo Thunder Trail, Santa Fe, New Mexico

Tuesday Evening, May 31, 2011 *(By Invitation Only)*

6:00 Opening Session – Committee Gathering and Reception

6:30 Director's Welcome and Committee Charge ....Duncan W. McBranch
PADSTE; Deputy Principle Associate Director, Science, Technology & Engineering

7:00 ADEPS Welcome and Expectations..Susan J. Seestrom
ADEPS; Associate Director, Experimental Physical Sciences

7:15 Executive Session *(Committee Only)*.. Gary S. Was
Committee Chair

Purpose:	**Annual Capability Review**	Classification Level:	Unclassified/Classified
Institutional Host(s):	Susan J. Seestrom, ADEPS, 505-665-4454	Dress:	Business/Business Casual
Technical Host(s)	Wendy R. Cieslak, MST-DO, 5-1535	Revised:	5/18/11 (jms)
Protocol POC:	Peggy S. Vigil, CGA-GAO, 7-8448 or cell 699-2195		
Catering:	ARAMARK		
LANL Update:	505-667-6622 or 1-877-723-4101: Provided information about changes in the Laboratory schedule (i.e., closings or delays) Protocol Office will adhere to all weather delays/closings.		

Agenda

Materials Capability Review

Wednesday, June 1, 2011

7:00 Meet visitors in lobby of the Hilton @ Buffalo Thunder .... Peggy S. Vigil
CGA-GAO: PADSTE, Protocol

7:05 Travel to LANL Badge Office, Bus leaves the hotel *(promptly @ 7:05)* LANS Taxi Service

7:30 Badge Office Processing .... Peggy S. Vigil

8:00 Walk to J. Robert Oppenheimer Study Center (breakfast provided for committee)

J. Robert Oppenheimer Study Center
TA-3, Bldg. 207, Jemez/Cochiti Room

8:05 Committee Photo .... Robert W. Kramer
ADEPS Communications Team

8:20 Welcome and Logistics .... Susan J. Seestrom
ADEPS; Associate Director, Experimental Physical Sciences

8:30 Overview of the Materials Capability at Los Alamos .... Wendy R. Cieslak
MST-DO; Division Leader, Materials Science & Technology

9:15 Materials for the Future: *Towards an Implementation Plan for the Materials Strategy*
.... Antoinette (Toni) Taylor
MPA-DO; Division Leader, Materials Physics & Applications

9:55 MaRIE Update .... T. Mark McCleskey
MPA-MC; Group Leader, Materials Chemistry

10:25 Break

10:40 LDRD-DR Review: Predictive Design of Noble Metal Nanoclusters .... Jennifer Martinez
MPA-CINT; Scientist, Center for Integrated Nanotechnologies

11:40 LDRD-DR Discussion led by .... William Priedhorsky
LDRD-PO; Program Director, Laboratory Directed Research & Development
Including Susan Seestrom, Cathy Cleland, Wendy Cieslak, and Toni Taylor

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J. Robert Oppenheimer Study Center
TA-3, Bldg. 207, Jemez/Cochiti Room

12:00 Working Lunch with Early Career Staff and Postdocs *(By Invitation Only)*
(San Ildefonso Conference Room)

1:30 **Materials in Radiation Environments** Overview .... Jack S. Shlachter
T-DO; Deputy Division Leader, Theoretical Division

2:00 Center for Materials at Irradiation and Mechanical Extremes (CMIME) Michael A. Nastasi
MPA-CINT; Laboratory Fellow, Center for Integrated Nanotechnologies

2:30 Research Activities at the Ion Beam Materials Laboratory (IBML) .... Yongqiang Wang
MST-8; Scientist, Structure/Property Relations

3:00 Discussion and Questions .... Jack S. Shlachter
T-DO; Deputy Division Leader, Theoretical Division

3:15 Posters Presentations .... Various Staff
Poster Titles and Presenters: See Appendix A, Page 8

3:30 Poster Session
Santa Clara Gallery

4:30 Executive Session *(Committee Only)*.. Gary S. Was
Committee Chair

5:30 Travel to the O Eating House Restaurant *(Committee Only)*.............................. Taxi Service
86 Cities of Gold Rd., Santa Fe, NM, Phone 505-455-2000

6:00 Hosted working Dinner (*By Invitation Only*)
Fukushima Reactor Studies .... R. Dasari Venkateswara (DV Rao)
D-DO; Division Leader, Decision Applications

8:30 Travel to the Hilton @ Buffalo Thunder .... Taxi Service

Materials Capability Review

Thursday, June 2, 2011

7:00 Meet visitors in lobby of the Hilton @ Buffalo Thunder .... Peggy S. Vigil
CGA-GAO: PADSTE, Protocol

7:05 Travel to LANL Study Center, Bus leaves the hotel *(promptly @ 7:05)* .... Taxi Service

J. Robert Oppenheimer Study Center
TA-3, Bldg. 207, Jemez/Cochiti Room

7:30 Voice of the Customer *(By Invitation Only)* (breakfast provided for committee)
(San Ildefonso Conference Room)

9:00 **Materials for Nuclear Energy (Focus on Actinides)** Overview .... David F. Teter
MST-DO; Deputy Division Leader, Materials Science & Technology

9:30 Lower Length Scale Simulations of Fission Gas and Thermal Transport in Oxide Nuclear Fuel .... Christopher R. Stanek
MST-8; Scientist, Structure/Property Relations

10:00 Development of Experimental Techniques to Augment Theory and Simulation of Thermal Transport in Oxide Nuclear Fuels .... Andrew T. Nelson
MST-7; Scientist, Polymers & Coatings

10:30 Discussion and Questions .... David F. Teter
MST-DO; Deputy Division Leader, Materials Science & Technology

10:45 Poster Presentation .... Various Staff
Poster Titles and Presenters: See Appendix B, Page 9

11:00 Poster Session
Santa Clara Gallery

Purpose:	**Annual Capability Review**	Classification Level:	Unclassified/Classified
Institutional Host(s):	Susan J. Seestrom, ADEPS, 505-665-4454	Dress:	Business/Business Casual
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Protocol POC:	Peggy S. Vigil, CGA-GAO, 7-8448 or cell 699-2195		
Catering:	ARAMARK		
LANL Update:	505-667-6622 or 1-877-723-4101: Provided information about changes in the Laboratory schedule (i.e., closings or delays) Protocol Office will adhere to all weather delays/closings.		

J. Robert Oppenheimer Study Center
TA-3, Bldg. 207, Jemez/Cochiti Room

12:00 Panel Discussion and Lunch *(Committee and Panel Only)*
(San Ildefonso Conference Room)
The Relationship of MaRIE and LANSCE to the Material Strategy .... Panelists:
Wendy R. Cieslak, facilitator
Kurt F. Schoenberg
John L. Sarrao
Antoinette (Toni) Taylor

1:30 **Materials for Clean Energy** Overview .... David E. Watkins
MPA-DO; Deputy Division Leader, Materials Physics & Applications

2:00 Center for Advanced Solar Photophysics (CASP) .... Victor I. Klimov
C-PCS; Laboratory Fellow, Chemistry, Physical Chemistry & Applied Spectroscopy

2:30 Materials Development in the LANL Fuel Cell Program .... Rodney L. Borup
MPA-11; Program Manager, Sensors & Electrochemical Devices

3:00 Discussion and Questions .... David E. Watkins
MPA-DO; Deputy Division Leader, Materials Physics & Applications

3:15 Poster Presentations .... Various Staff
Poster Titles and Presenters: See Appendix C, Page 10

3:30 Poster Session
Santa Clara Gallery

4:30 Executive Session *(Committee Only)*

6:00 Travel to the Hilton @ Buffalo Thunder .... Taxi Service

Friday, June 3, 2011

LANSCE
TA-53, Building 1, Rosen Auditorium

7:30 Committee Members Arrive & Finalize Tour Access/Logistics .... Gary S. Was
(breakfast provided for committee) *Committee Chair*

7:45 **LANSCE** Overview .... Kurt F. Schoenberg
ADEPS; Deputy Associate Director, Experimental Physical Sciences and LANSCE Director

8:15 Travel to the Lujan Center.. Taxi Service

8:30 LANSCE Tour Begins -Lujan Center .... Alan J. Hurd
LANSCE-LC; Group Leader, Lujan Center

9:00 Travel to pRAD

9:10 LANSCE Tour –pRAD .... Alexander (Andy) Saunders
P-25; Deputy Group Leader, Subatomic Physics

9:40 Travel: Cleared to Building1; Uncleared to Injector Building

Cleared Committee Members Split Off for Classified Session shown in red
Uncleared Committee Members Session shown in black

SECURITY NOTICE: Electronics, including cell phones, two-way pagers, PDA's (Blackberry, Palm Pilot, etc.), laptop computers, thumb-drives, cameras, etc. are NOT allowed in cleared Laboratory areas. It is suggested that visitors going behind the security fence leave all personal belongings in vehicles or hotel rooms or they will be subject to a complete search to include coats, briefcases, etc.

9:50 *Location: TA-53-1-D143*
Classified pRAD Session .... Christopher Morris
P-25; Laboratory Fellow, Subatomic Physics

10:10 *Classified Poster Session*.. Various Staff
Poster Unclassified Titles and Presenters: Listed with Theme, pages 8 and 9

LANCSE Tour

9:50 LANSCE Tour – Injector Building .... John L. Erickson
AOT-DO; Division Leader, Accelerator Operations & Technology

10:20 Return to Building 1 .... Taxi Service

10:30 Committee Rejoins for Executive Session in A234 *(Committee Only)* .... Gary S. Was
Committee Chair

11:30 Lunch *(Closed Session)* .... Terry C. Wallace Jr.
PADSTE; Principle Associate Director, Science, Technology & Engineering

12:40 Committee Members to the Rosen Auditorium

Rosen Auditorium
TA-53, Building 1

12:45 Formal Outbrief .... Gary S. Was
Committee Chair

2:00 Committee Members Depart

APPENDIX A

***MATERIALS IN RADIATION ENVIRONMENTS POSTER SESSION**, Wednesday, June 1, 2011*

Irradiation Environment in the Proposed Materials Test Station .... Eric J. Pitcher
LANSCE-DO, LANSCE

LANSCE: Nuclear Physics and Materials Science ....John L. Ullmann
LANSCE-NS, Neutron & Nuclear Science

The Role of Grain Boundaries in Modifying Radiation Damage Evolution in Simple Metals
.... Blas P. Uberuaga
MST-8, Structure/Property Relations

Atomistic Modeling and Experimental Studies of the Shock Response of Cu/Nb Nanolayered Composites ....Ruifeng Zhang
T-3, Fluid Dynamics and Solid Mechanics

Irradiation Damage Effects in Ceramic Oxides Induced by High Temperature and High Fluence Ion Beam Irradiation...Igor O. Usov
MST-7, Polymers & Coatings

IBML Proton Calibration Results of the SABRS Energetic Particle Subsystems
....Jane M. Burward-Hoy
ISR-1, Space Science & Applications

Effects of Radiation Environments on Long-Term Aging of Weapon Materials (U)
.... Jennifer A. Lillard
MST-6, Materials Technology-Metallurgy
Presented in Classified Session June 3, 2011

Advanced Materials Development and Testing for Fuel Cycle Research and Development Program
.... Osman Anderoglu
MST-8, Structure/Property Relations

Influence of Interstitials or Vacancies on Grain Boundary Sliding Processes in bcc Tungsten
.... Art F. Voter
T-1, Physics and Chemistry of Materials

APPENDIX B

***MATERIALS FOR NUCLEAR ENERGY (FOCUS ON ACTINIDES) POSTER SESSION**, Thursday, June 2, 2011*

Sintering of Mixed-Oxide Fuel Pellets .... Jeremy N. Mitchell
MST-16; Nuclear Materials Science

Plutonium Science and Research Strategy: Use of Special Isotopes – 242Pu .... Dom S. Peterson
MST-7, Polymers & Coatings

Process Modeling of Plutonium and Uranium Casting .... David A. Korzekwa
MST-6, Materials Technology-Metallurgy

LEU U-10Mo Research Reactor Fuel Development and Scale Up............... David E. Dombrowski
MST-6, Materials Technology-Metallurgy

Residual Stresses in Aluminum Clad Uranium-10wt%molybdenum Fuel Plates
.... Donald W. Brown
MST-8, Structure/Property Relations

Chemical Segregation of U-10Mo Fuel Foils during Simulated Bonding Cycles Using Neutron Diffraction .... Sven C. Vogel
LANSCE-LC; Lujan Center

Radioparagenesis: Robust Nuclear Waste Form Design and Novel Materials Discovery
.... Boris Dorado
MST-8, Structure/Property Relations

Fissionable Scintillators for Measuring Neutron Flux .... Ernst I. Esch
N-1, Safeguards Science and Technology

Material Attractiveness in Nuclear Fuel Cycles (U) .... Charles G. Bathke
D-5, International & Nuclear Systems Engineering
Presented in Classified Session June 3, 2011

APPENDIX C

***MATERIALS FOR CLEAN ENERGY POSTER SESSION**, Thursday, June 2, 2011*

Colloidal Nanomaterials for Light Harvesting Applications.....................................Anshu Pandey
C-PCS; Physical Chemistry & Applied Spectroscopy

Nanocrystals .... John T. Stewart, IV
C-PCS; Physical Chemistry & Applied Spectroscopy

Assessment of Silicon Nanowire Architecture for PV Application .... Shadi Dayeh
MPA-CINT, Center for Integrated Nanotechnologies

Controlling Electrode Morphology to Improve Fuel Cell Performance and Durability
.... Cynthia F. Welch
MST-7, Polymers & Coatings

Nanoscale Ceramic Catalyst Support Synthesis .... Eric L. Brosha
MPA-11, Sensors & Electrochemical Devices

Membrane Materials for Energy Applications .... Katherine A. Berchtold
MPA-MC; Materials Chemistry

Chemical H2 Storage Research at LANL.. Benjamin L. Davis
MPA-MC; Materials Chemistry

Comparative Studies of Vortex Physics in Oxide, Iron-arsenide and MgB2 Superconductors
....Leonardo Civale
MPA-STC; Superconductivity Technology Center

Ions to Wires: The Science of High Tc Superconducting Wires...................... Terry G. Holesinger
MPA-STC; Superconductivity Technology Center

CHARTER

For the 2011 Los Alamos National Laboratory Materials Capability Review Committee

The 2011 "Capability Review" process at LANL significantly differs from the Division reviews of prior years. The Capabilities being reviewed (some 4-8 per year) are deliberately chosen to be crosscutting over the Laboratory and therefore will include not only several experimental, theoretical and simulation disciplines but also contributions from multiple line organizations. This approach is consistent with the Laboratory organizational structure, focusing on agile and integrated capabilities applied to present national security missions but also nurtured to be available for rapid application to future missions.

The overall intent is that the Committee **Assess** the quality of the science, engineering, and technology identified in the agenda, and **Advise** the LANS Board of Governors and Laboratory Management.

Specifically, Committees will:

- Assess the quality of science, technology and engineering within the capability in the areas defined in the agenda. Identify issues to develop or enhance core competencies in this capability.

- Evaluate the integration of this capability across the Laboratory organizations that are listed in the agenda in terms of joint programs, projects, proposals, and/or publications. Describe the integration of this capability in the wider scientific community using the recognition as a leader within the community, ability to set research agendas, and attraction and retention of staff.

- Assess the relevance of this capability's science, technology and engineering contributions to current and emerging LANL programs, including Nuclear Weapons, Global Security, and Energy Security.

- Advise the Laboratory Director/Principal Associate Director for Science, Technology and Engineering on the health of the capability including the current and future (5 year) science, technology and engineering staff needs, mix of research and development activities, program opportunities, environment for conducting science, technology and engineering.

- An additional specific charge for the Materials Capability Review is to assess the Los Alamos Laboratory Directed Research and Development (LDRD) project titled, "Predictive Design of Noble Metal Nanoclusters" using the criteria performance, quality, and relevance for the current status of the project. Provide advice on the future directions and opportunities for the project.

Instructions for the Los Alamos National Laboratory Fiscal Year 2011 Capability Reviews

Introduction

Los Alamos National Laboratory (LANL) uses external peer review to measure and continuously improve the quality of its science, technology and engineering (STE). LANL uses capability reviews to assess the STE quality and institutional integration and to advise Laboratory Management on the current and future health of the STE. Capability reviews address the STE integration that LANL uses to meet mission requirements. STE capabilities are define to cut across directorates providing a more holistic view of the STE quality, integration to achieve mission requirements, and mission relevance. The scope of these capabilities necessitate that there will be significant overlap in technical areas covered by capability reviews (e.g., materials research and weapons science and engineering). In addition, LANL staff may be reviewed in different capability reviews because of their varied assignments and expertise. LANL plans to perform a complete review of the Laboratory's STE capabilities (hence staff) in a three-year cycle. The principal product of an external review is a report that includes the review committee's assessments, commendations, and recommendations for STE.

The Capability Review Committees have the primary responsibility of assessing the Laboratory's STE quality through the topics selected for the review. The Committees will also evaluate the integration of STE within the capability. Capability reviews are about the quality of STE, but it is difficult to separate the STE from the Laboratory missions and programs. The Committees are charged with evaluating the relevance of the STE towards the Laboratory's missions and programs, but capability reviews are not program reviews and are not expected to perform an in-depth examination of programs. Finally Capability Review Committees provide advice to Laboratory Management on STE issues identified during the review. The assessments and advice are documented in reports prepared by the Capability Review Committees that are delivered to the Director and to the Principal Associate Director for Science, Technology and Engineering (PADSTE). Laboratory Management will use this report for STE assessment and planning. The report is also provided to the Department of Energy (DOE) as part of LANL's Annual Performance Plan and to the Los Alamos National Security (LANS) LLC's Science and Technology Committee (STC) as part of its responsibilities to the LANS Board of Governors.

LANL has defined fourteen STE capabilities. Table 1 lists the seven STE capabilities that LANL Management (Director, PADSTE, technical Associate Directors) have identified for review in Fiscal Year (FY) 2011. The FY 2011 capability reviews must be **completed by June 30, 2011** to allow sufficient time for the reports and results to be incorporated into the 2011 STE evaluations by the Department of Energy National Nuclear Security Agency and the LANS LLC STC for the LANS Board of Governors.

These instructions identify responsibilities and provide guidance to those organizing, participating in and performing capability reviews at LANL in FY 2011. These instructions have been refined based on experiences with capability reviews from 2007 - 2010. Any questions or comments on these instructions should be directed to the Program Manager for the LANL Science and Technology Base Programs Peer Review and Metrics Office at stbprm-admin@lanl.gov or 505-667-7824.

Table 1. FY 2011 LANL science, technology and engineering capability reviews, organizing associate director (AD), and Los Alamos National Security, LLC Science and Technology Committee point-of-contact (STC POC).

Capability	Organizing AD	STC POC/co-POC
Advanced Manufacturing	ADSMS	Peddicord/Benz
Biosciences	ADCLES	Colvin/Segalman
Information and Knowledge Sciences	ADTSC	Long/Karin
Materials	ADEPS	Navrotsky/Bercaw
Nuclear Engineering and Technology	ADTIR	Peddicord/Karin
Sensors, Remote Sensing and Sensor Systems	ADCLES	Gehrels/Beckwith/Vogt
Weapons Science and Engineering	ADW	Rosner/Peddicord

Roles and Responsibilities

LANL Director/PADSTE

1. Determines capabilities to be reviewed and review schedule.
2. Appoints an Organizing AD (OAD) for each capability review.
3. Works with the OAD to create a specific charge for each capability review.
4. Participates in Capability Review Committee member selection as needed.
5. In conjunction with the STC Chair approves the Capability Review Chair and members.
6. Invites the Capability Review Chair and members.
7. Hosts the Capability Review Chairs meeting before the review cycle starts.
8. Provides the charge to the Capability Review.
9. Attends executive session at closeout.
10. Ensures report is delivered by requested deadline.
11. Provides the Capability Review report to the STC Chair for distribution to the STC.
12. Addresses the Capability Review recommendations through the PADSTE Management Review Board assigning actions and resources. The PADSTE will determine if the actions are tracked through the LANL performance tracking system.
13. Incorporates review recommendations and issues into LANL STE planning and assigns actions.
14. Provides summary response for all capability reviews to Capability Review Committee Chairs and to the STC Chair.

LANS, LLC STC Chair (STC Chair)

1. Appoints STC members to serve as the point-of-contact (POC) and co-point-of-contact (co-POC) for each Capability Review Committee.
2. In conjunction with the LANL Director/PADSTE, approves the Capability Review Committee Chair and members.
3. The STC Chair or the Vice Chair may attend any Capability Review meeting as observers. Observers may participate in discussions and attend all sessions, but may not participate in drafting the report.

LANS, LLC STC Point of Contact (POC) / co-Point of Contact (co-POC)

1. Working with the Organizing AD (OAD), compiles a list of potential Capability Review Committee members based on input from the Laboratory, University of California (UC), and STC members. (UC Office of the President (UCOP) collects input from UC, including from the Academic Senate). At least one and a maximum of two LLNL staff members and at least one University of California faculty member are to be named to each Capability Review Committee. The POC does not count for either of these requirements.
2. Working with the OAD, prioritizes the list of potential Capability Review Committee Chairs and members. The list is given to the PADSTE and the STC Chair for approval.

3. The POC or OAD, as appropriate, contacts the recommended Capability Review Chair to ask if the person will serve; the PADSTE officially invites the Chair.
4. Reviews the prioritized Capability Review Committee membership candidate list with the selected Capability Review Chair and OAD; the STC Chair and PADSTE are notified of any changes.
5. Works with the Capability Review Committee Chair and OAD to identify potential dates for the Capability Review meeting.
6. With dates in hand, the POC, OAD, or Capability Review Committee Chair, as appropriate, contacts the proposed Capability Review Committee members regarding their willingness to serve until approximately 9 members, including the Chair, are enlisted. At least one and a maximum of two LLNL staff members and at least one University of California faculty member are to be named to each Capability Review Committee. The Capability Review organizing team (i.e., OAD, Capability Review Committee Chair, and POC) maintains contact during this step and consults if issues arise.
7. Participates in developing the Capability Review meeting agenda with the OAD and Capability Review Committee Chair.
8. Attends the Capability Review Chairs meeting hosted by the PADSTE before the review cycle starts.
9. Attends the Capability Review meeting as an ex-officio member, participates as a full member, including attendance at executive sessions, but does not participate in drafting the report.

LANL Organizing Associate Director (OAD)

1. Coordinates with LANL Director/PADSTE and with ADs who contribute to the capability to develop the Capability Review scope.
2. Works with the POC to prioritize the list of potential Capability Review Committee Chairs and members. The list is given to the PADSTE and the STC Chair for approval.
3. The OAD or POC, as appropriate, contacts the recommended Capability Review Committee Chair to ask if the person will serve; the PADSTE officially invites the Chair.
4. Reviews the prioritized Capability Review Committee membership candidate list with the selected Capability Review Committee chair and POC; the STC Chair and PADSTE are notified of any changes.
5. Works with the Capability Review Committee Chair and POC to identify potential dates for the Capability Review meeting.
6. With dates in hand, the OAD, POC, or Capability Review Committee Chair, as appropriate, contacts the proposed Capability Review Committee members regarding their willingness to serve until approximately 9 members, including the Chair, are enlisted. At least one and a maximum of two LLNL staff members and at least one University of California faculty member are to be named to each Capability Review Committee. The Capability Review organizing team (i.e., OAD, Capability Review Committee Chair, and POC) maintains contact during this step and consults if issues arise.

7. Participates in developing the review agenda with the POC and Capability Review Committee Chair.
8. Identifies Los Alamos Laboratory Directed Research and Development (LDRD) project to be reviewed for the current year. The LANL LDRD Office can assist in project identification.
9. Attends the Capability Review Chairs meeting hosted by the PADSTE before the review cycle starts.
10. Compiles and sends background information to the Capability Review Committee before the review.
11. Provides logistics for the Capability Review meeting, including meeting rooms, necessary security for classified sessions, etc.
12. Works with the Capability Review Committee Chair to maintain review agenda and schedule.
13. Provides additional information requested by the Capability Review Committee.
14. Addresses Capability Review recommendations assigned by Director/PADSTE.

Capability Review Committee Chairperson

1. Reviews the prioritized Capability Review Committee membership candidate list with the selected OAD and POC; the STC Chair and PADSTE are notified of any changes.
2. Works with the organizing AD and POC to identify potential dates for the Capability Review meeting.
3. With dates in hand, the OAD, POC, or Capability Review Committee Chair, as appropriate, contacts the proposed Capability Review Committee members regarding their willingness to serve until approximately 9 members, including the Chair, are enlisted. At least one and a maximum of two LLNL staff members and at least one University of California faculty member are to be named to each Capability Review Committee. The Capability Review organizing team (i.e., OAD, Capability Review Committee Chair, and POC) maintains contact during this step and consults if issues arise.
4. Participates in developing the Capability Review meeting agenda with the POC and OAD. The Chair ensures sufficient executive time in the agenda for committee discussions.
5. Attends a meeting of the Capability Review Committee Chairs hosted by the PADSTE before the review cycle starts.
6. Distributes information about the meeting to Capability Review Committee members as necessary.
7. Presides over the review by keeping to the agenda, managing deliberations of the Capability Review Committee, and assigning tasks to Capability Review Committee members as appropriate.
8. Prepares and leads executive out-brief to the Director/PADSTE.
9. Provides Capability Review report to the LANL Director/PADSTE within 30 days of the review.

Capability Review Committee Members

1. Attend review and complete tasks assigned by Capability Review Committee Chair.
2. Provide unbiased and objective evaluation of the topics within the capability being assessed.
3. Provide written material to Capability Review Committee Chair with sufficient time to meet schedule.

Assessment

The evaluation of designated topics must address the following two criteria:

1) Comparison to peers -- State how the work compares to similar or related work conducted by others.
2) Sustainability -- State the extent to which the reviewed activities strengthen or weaken LANL capabilities. How does the activity/contribution build core competencies or other resources that contribute to the vitality of the capability and the long-term vigor of the Laboratory and its ability to meet the needs of the nation?

Laboratory Directed Research and Development Assessments for FY 2011 Capability Reviews

The Director/PADSTE will charge each Capability Review Committee to assess a single Laboratory Directed Research and Development (LDRD) project related to the capability. Many of the three-year LDRD projects will be nearing completion enabling the Capability Review Committee to assess the quality of the work performed and provide guidance on the programmatic development of the project.

The Capability Review Committee is requested to prepare an assessment (one to two pages) of the LDRD project that will be included in the Capability Review report and in the LDRD Annual Report to the DOE. The selected LDRD project will be included in the agenda as a presentation, and the Capability Review Committee will be asked to assess the project using the following criteria:

1. Quality: Are the science and technology results of high quality compared to national and international peers?
2. Performance (project execution): Is the project making good progress against its milestones? Is it well-conceived and executed?
3. Relevance: Is the project continuing to support the strategic directions of the Laboratory?
4. Leadership: Are the results of the project defining R&D directions for the broader community?

The LDRD office will provide a guidance sheet to assist the committee in evaluating the project. The use of the guidance sheet will allow a common framework for LDRD project evaluation with other reviews that LANL performs of its LDRD projects. The committee cannot fully implement the guidance because the capability review duration is shorter than allowed for other reviews, but the guidance does indicate some key concepts that we would like to see addressed.

The Capability Review Committee's advice on future directions and opportunities for the project is requested.

Briefing management

At the end of it's meeting, the Capability Review Committee will brief its findings to LANL Management. Attendance at this briefing, other than senior management (Director/PADSTE), remains at the discretion of the Capability Review Committee Chair and the Director/PADSTE. The out-brief should provide executive style highlights of the assessments and advice for the capability. The Capability Review Committee should prioritize its assessment and advice for the out-briefing (and the report). Specifically, the Capability Review Committee should deliberate in its executive session to identify and prepare for presentation:

1) 3 to 7 most notable contributions observed in the review, and
2) 3 to 7 most important "actionable" recommendations.

Each of these components of the out-briefing should be presented in order of decreasing importance or significance (highest, next to highest, etc.). The rationale behind prioritization is to engage the wisdom and experience of the Capability Review Committee to identify the true pinnacles and the most significant challenges. It is the distinctiveness of the greatest achievements and the magnitude of the greatest challenges that characterize the excellence of an organization/program.

By prioritizing a limited number of items, Capability Review Committees are able to focus their feedback and enable meaningful follow up by LANL Management. A template containing recommended content for the out-briefing can be found in the Appendix of this document.

Preparing the Capability Review report

The Capability Review Committee must submit its assessment and advice via written report. The final copy is due to the Program Manager in the Science and Technology Based Programs Peer Review and Metrics Office within 30 working days of the end of the Capability Review meeting. The Capability Review Committee Chair is responsible for delegating writing assignments, coordinating inputs, editing the final document, and submitting it.

A suggested report template can be found in the Appendix of this document. The template includes abstracts of the areas to be assessed and headings delineating the areas in which specific advice has been requested. The assessment of the LDRD project can follow the same format, but the three criteria (performance, quality, and relevance) that were identified in these instructions need to be addressed.

Appendix

Capability Review Out-Brief Template

Acknowledgement and Recognition
- Opening remarks
- Feedback on execution of review

Assessment of Topics in the Agenda
- Comparison to peers, mission/program relevance, integration
- Assessment of LDRD project

Prioritized Conclusions
- Top 3 to 7 Capability Review Committee "actionable" recommendations
- Top 3 to 7 most notable science, technology and engineering contributions

Special topics
- Any needs for additional information or meetings
- Topics of enduring interest beyond the annual review cycle (e.g. from prior reviews)
- Improvements in capability review process

Capability Review Committee Report Template

Title
Table of Contents
Executive Summary
Introduction
Assessment

- Review Elements (*directly from agenda)*
- Review element 1
  - Scope of the review
    - *Can use pre-written element description (single 50-300 word abstract written by LANL contributor summarizing goal of contribution and key results)*
  - Analysis (*in terms of one or more of these 4 facets)*
    - Approach
    - Implementation
    - Results
    - Impact of work
  - Assessment
    - Comparison to peers
    - Sustainability
- Review element 2
- Review element n
- Review of LDRD project
  - Performance
  - Quality
  - Relevance
  - Capability Review Committee's advice on future directions for the project

Prioritized Conclusions

- Top 3 to 7 most notable science, technology and/or engineering contributions (or other high performance indicators)
- Top 3 to 7 Capability Review Committee "actionable" recommendations

Acknowledgements
Appendices

- Capability Review Committee Meeting Agenda
- Roster of Capability Review Committee Members
- Additional inputs or documents used in assessment by Capability Review Committee

Overview of the Materials Capability at Los Alamos National Laboratory

W.R. Cieslak (MST-DO)

As one of the three foundational pillars of Los Alamos, the Materials Capability covers an exceptional breadth and depth. Materials science and engineering is instrumental to enabling all the Laboratory's missions and staff all across the Laboratory engage in materials research. The Los Alamos Materials Capability is comprised of innovative research and development at the boundaries of chemistry, physics, theory, and materials science that translates fundamental discovery to materials lifecycle applications in topics central to our mission, such as actinide science.

The purpose of this presentation is to orient the committee to this capability and what is being covered over the course of the review. As such, the presentation covers four main topics:

1) Summarize the three-year rotational cycle of review topics and the agenda for this meeting.
2) Reprise the recommendations from last year's Materials Capability Review and the changes that have occurred in response.
3) Discuss metrics on the institutional demographics for the capability, including budgets, people and publications.
4) Point out a few technical highlights from the past year that are outside of what is being covered in this year's review.

Overview of the Materials Capability at Los Alamos National Laboratory

Materials Capability Review 2011

Wendy R. Cieslak

Division Leader

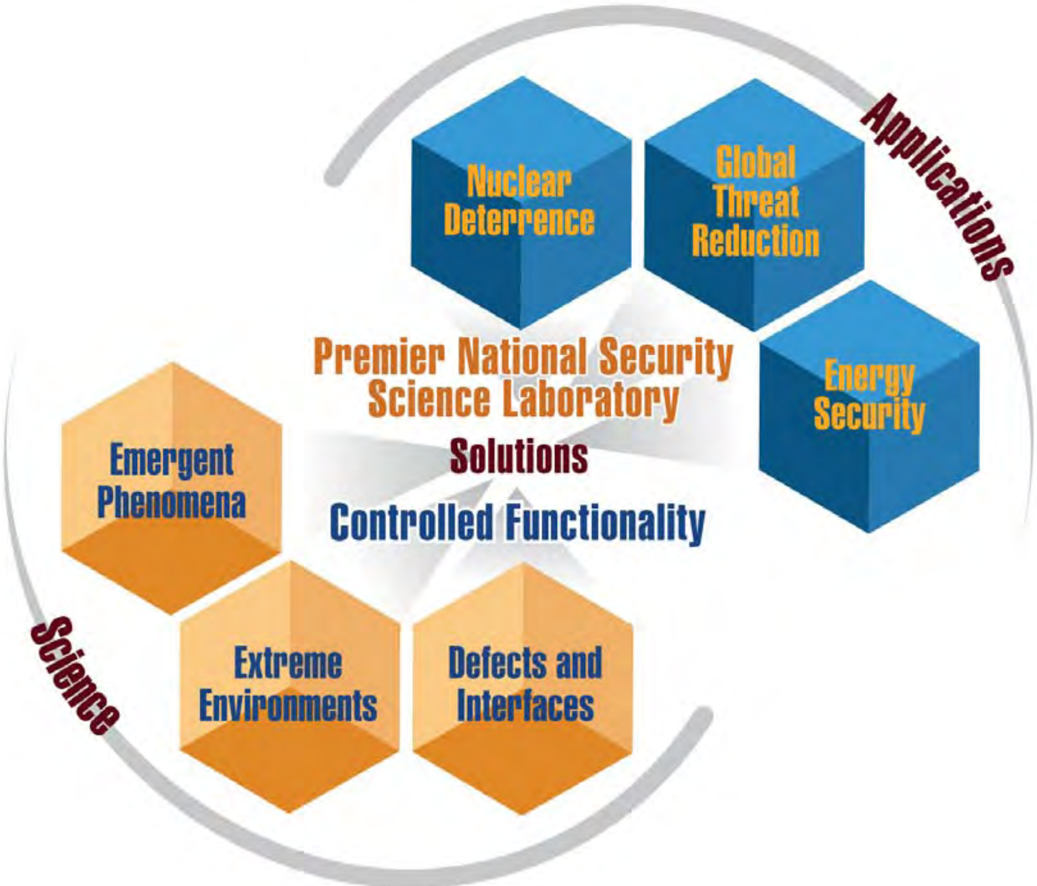
Materials Science & Technology

UNCLASSIFIED

Presentation outline

- **2011 Topics**
- Response to 2010 recommendations
- Trends in metrics
- Technical highlights

We are organizing our Materials Capability Reviews based on the Materials Strategy



Crosscuts:

- **Making**
- **Measuring**
- **Modeling**

We are organizing our Materials Capability Reviews based on the Materials Strategy

	FY-10: Global Security	FY-11: Energy Security	FY-12: Nuclear Deterrence
Overview	▪ Materials Overview ▪ Materials Strategy ▪ MaRIE update	▪ Materials Overview ▪ Materials Strategy ▪ MaRIE update	▪ Materials Overview ▪ Materials Strategy ▪ MaRIE update
Emergent phenomena	CINT/Nanoscience	Materials for Clean Energy	Condensed Matter Physics/NHMFL
Defects and Interfaces	Electronic/ Photonic Materials	Materials for Nuclear Energy (Actinides)	Surface science/ corrosion
Extreme Environments	Materials Dynamics	Materials in Radiation Environments	HE/High Pressure
Programmatic	Materials for Global Security	- Deleted -	- Deleted -
Critical Issues	▪ CINT tour ▪ Pu strategy ▪ Actinide LDRD	• LANSCE tour • Nanoclusters LDRD	• PF4/CMRR tour • LDRD

2011 Materials Capability Review topics and presenters

- **Materials Vision (including panel discussion)**
  - Materials Strategy and Implementation*: Toni Taylor, Division Leader, MPA*
  - MaRIE update: *John Sarrao, Program Director, Office of Science & MaRIE*
- **Extreme Environments: Materials in Radiation Environments**
  - Lead: *Jack Shlachter, Deputy Division Leader, Theoretical (T)*
- **Defects and Interfaces: Mat'ls for Nuclear Energy (Actinides Focus)**
  - Lead: *Dave Teter, Deputy Division Leader, MST*
- **Emergent Phenomena: Materials for Clean Energy**
  - Lead: *Dave Watkins, Deputy Division Leader, MPA*
- **Critical Issues**
  - LANSCE Overview and Tours: *Kurt Schoenberg, Deputy Associate Director, ADEPS and Alex Lacerda, Deputy Division Leader, LANSCE*
  - 3rd year LDRD-DR review: "Predictive Design of Noble Metal Nanoclusters," *Jennifer Martinez, Scientist, MPA*

Your feedback on the performance of the Materials Capability is requested

- **Scientific Leadership as Evidenced by:**
  - Leadership in an international technical community
  - Publication of highly cited research results
  - Flux of innovative ideas and proposals
  - Hiring and training of next generation's leaders
- **Programmatic Impact**
- **Sustaining a *Cross-Laboratory* Materials Community**
- **Direction for the Future**
  - Progress on implementing the Materials Strategy
  - LANSCE and the path to MaRIE
  - Recapitalization and facilities revitalization

Presentation outline

- 2011 Topics
- **Response to 2010 recommendations**
- Trends in metrics
- Technical highlights

Major concerns/recommendations from 2010 were:

(a) to proceed quickly to develop an actinide sciences vision that incorporates both weapons and civilian nuclear energy applications;

(b) that the management seek a better balance between program execution and risk management;

(c) to address the growing need to bring in additional mid-career talent to allow the lab to stay ahead of upcoming;

(d) to move rapidly to establish a plan to sustain the now state-of-the-art electron microscopy facility;

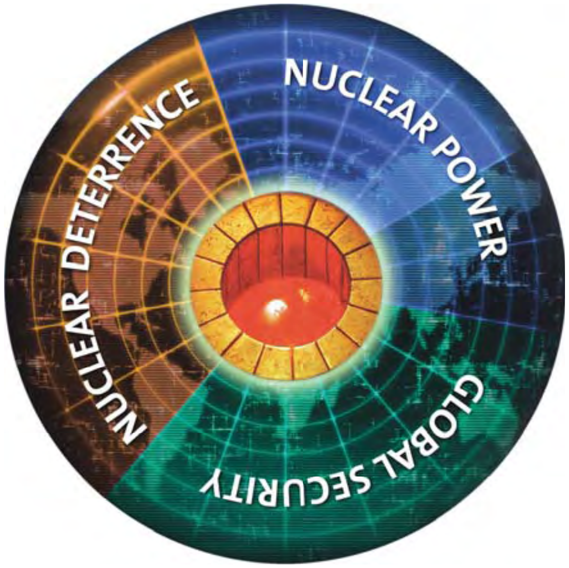
(e) to continue to define and refine first experiments that demonstrate/ utilize the unique capabilities of MaRIE that do not currently exist.

Actinide sciences vision that incorporates both weapons and civilian nuclear energy applications (1)

- **<u>LANL Pu Strategy</u> defines a road map for Pu research and development efforts that share a common underpinning in nationally important missions**

  Nuclear Deterrence Nuclear Power Global Security

- Includes the Laboratory's enduring roles in Pu science, and the technical and programmatic relationships between capabilities that support Pu science
- Defines scientific themes and resources to maintain LANL's Pu science leadership role
- Personnel, facility, instrumentation investments, and Strategic partnerships required to meet program objectives
- An implementation plan, including formulation of institutional investment strategies impacting Pu science



Actinide sciences vision (2) that crosscuts all three LANL mission elements

	nuclear deterrence	sustainable nuclear energy	reducing global nuclear threats
science			
	• electronic structure • phase stability • dynamic behavior • surface reactions • chemical separations • detection and analysis	• electronic structure • phase stability • surface reactions • chemical separations • nuclear energy systems • detection and analysis • environmental behavior	• dynamic behavior • surface reactions • chemical separations • nuclear energy systems • detection and analysis • environmental behavior
infrastructure			
	• Pu in LDRD program • science infrastructure • Small scale R&D w Pu-242 • Classified publ databases	• Pu in LDRD program • science infrastructure • Small scale R&D w Pu-242	• Pu in LDRD program • science infrastructure • Small scale R&D w Pu-242 • Classified matl databases
workforce			
	• Promote technical work • Education and training	• Promote technical work • Education and training	• Promote technical work • Education and training

Management seek a better balance between program execution and risk management

- **Small scale Pu science has picked up substantially**
  - Example: On track to complete 15 shots on 40 mm gun (8 executed to date) in FY11 as compared to 5 shots in FY10
  - Science sample requirements are reviewed at weekly TA-55 program meeting
- **The two-man rule is lifted for PF-4 non-laboratory spaces on 6/1**
- **MST has piloted a graded approach to satisfy facility work control requirements (e.g. daily pre-jobs)**
- **Mid-April news headline: "LANL Director: Time for NNSA to Ease up on Lab Oversight"**
- **Our success is making the news, too (Alpha Pu on Z)**

We need to continue to make progress for scientists across the Laboratory to feel relief

Management seek a better balance between program execution and risk management

- Small scale P… y
  - Example: O… 8 executed to date) in FY1…
  - Science sam… -55 program meeting
- The two-man … spaces on 6/1
- MST has pilo… ity work control requi…
- Mid April nev… NNSA to Ease up on Lab O…
- Our success… Z)

Plutonium experiments conducted at Sandia's Z Accelerator Facility

"The successful Z Machine experiment demonstrates our commitment to… the safety, security and effectiveness of a smaller stockpile without nuclear testing,"
Donald Cook, Defense Programs

We need to continue to make progress for scientists across the Laboratory to feel relief

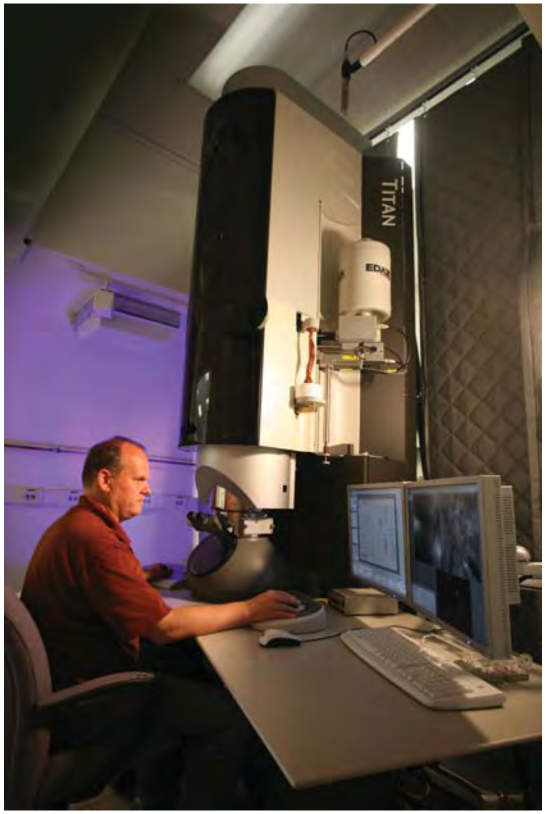
Growing need to bring in mid-career talent to stay ahead of upcoming opportunities

- **Hiring**
  - Two hires from LLNL (MST)
  - Attractive package offered in a timely manner; declined (MPA)
    - Early career scientist, hired externally, now fills the position
  - One re-hire from INL (T)
  - Strategic hire for biofuels (B)
- **Separations**
  - Attractive retention package offered; left for Argonne (MPA)
  - Couple leaving to be closer to family and dept. chair at U Akron (MST)
  - One considering department chair at U Tennessee (MST)
  - One loss to a University, others pending (T)
  - Two losses to Sandia (T and WX)

We need a holistic strategy; follow-up in metrics section

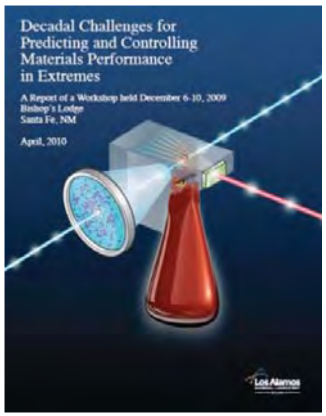
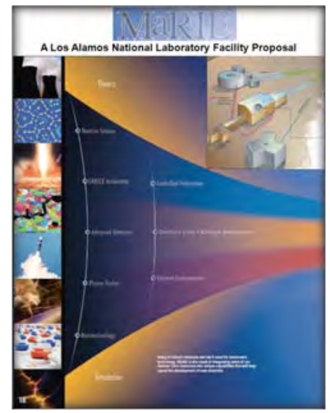
Need to rapidly establish a plan to sustain the now state-of-the-art electron microscopy facility

- **Challenge was balancing compliance with fiscal policy, equitable distribution of costs and an affordable user fee**
- **Examined several possible funding options:**
  - Direct Program Allocations
  - G&A
  - Org Support
  - Recharge model
- **PADSTE approved pursuing the recharge method – pending CFO approval effective Oct. 2011 (FY12)**
- **PADSTE also supported developing a 3-5 yr capital purchase investment plan**



Continue to define and refine first experiments that demonstrate/utilize the unique capabilities of MaRIE (1)

In response to the Dept's guidance, we are developing a pre-conceptual design proposal for MaRIE

Science Need	Facility Definition	Pre-conceptual Proposal
		
(2009)	(2010)	(2011)

Today

Current activities with NNSA and NE are key steps down the path:

LANSCE→ Linac Risk Mitigation → MTS → MaRIE

MaRIE (2): FY11 LDRD Reserve investments are accelerating science underlying first experiments

XFEL physics

- Coherence Effects in x-ray Diffraction Imaging — *Quinn Marksteiner/Barber*
- Control of XFEL-Radiation Focusing through Electron-Beam Manipulation — *Kip Bishofberger*

APS expts

- Synchrotron X-ray Laue Diffraction and Phase Contrast Imaging of Fe and Explosive Simulants under Shock Loading — *Shengnian Luo*
- In-Situ Probing Monitoring of Microstructure Evolution During Annealing of Radiation Damage with High Energy Synchrotron Xray Diffraction — *Donald Brown*
- Three Dimensional Quantification of Metallic Microstructures in the Presence of Damage — *Curt Bronkhorst*

Advanced pRad

- Achieving the Ultimate Spatial and Density Resolution of 800 MeV Proton Radiography — *Alexander Saunders*

Synthesis science

- Developing and synthesizing epitaxial nanocomposites with controlled defect landscapes and desired functionalities — *Quanxi Jia*
- Fluid Flow Imaging of Alloy Melts and In-situ Fundamental Solidification Experiments at Temperature Extremes — *Amy Clarke*
- Microstructure Analysis for Extreme Events: A Stochastic Modeling Framework for Microstructure Datasets — *John Bingert*

- Accelerating Materials Certification — *John Sarrao*

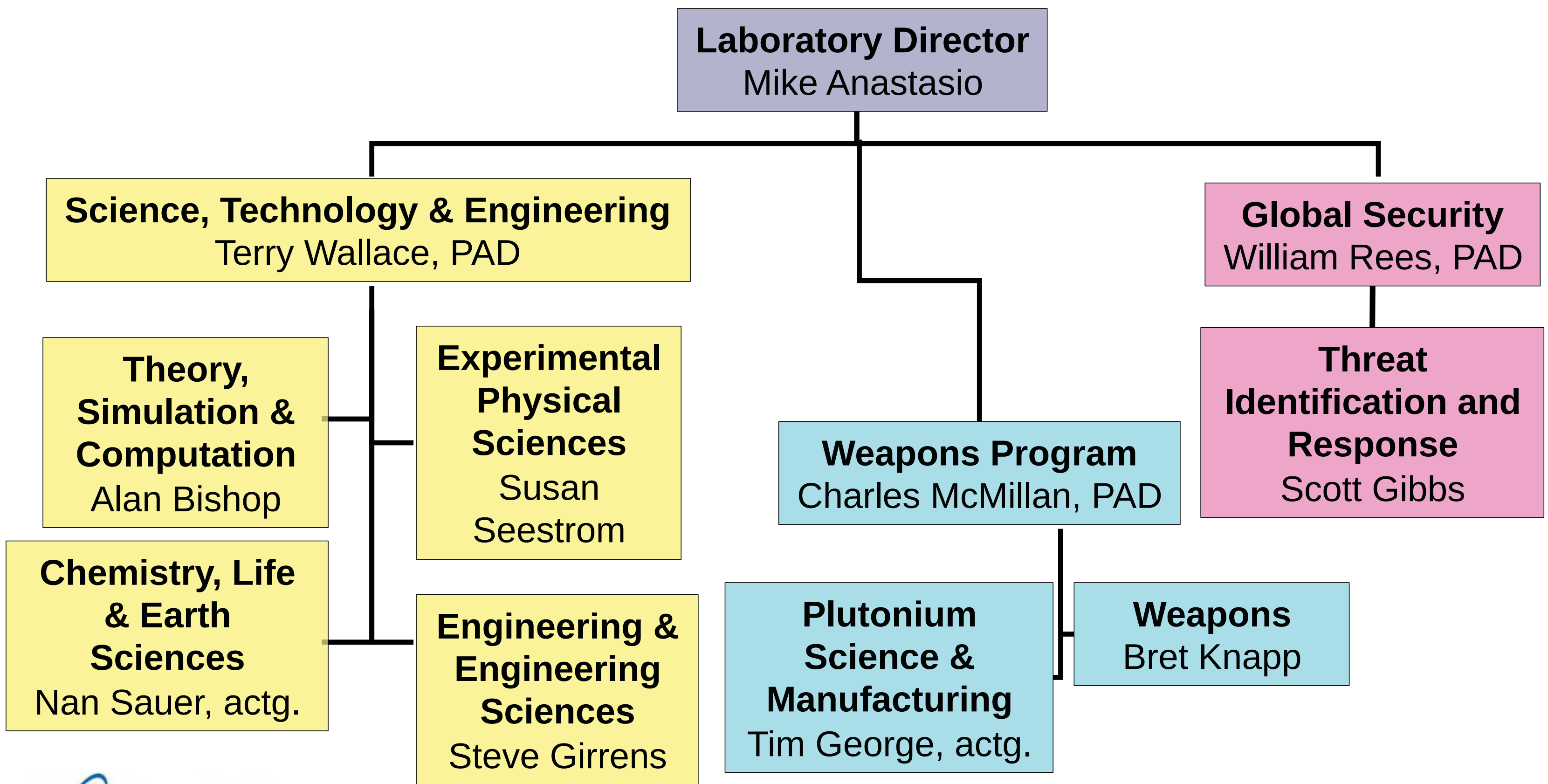
Minor concerns/observations from 2010

- The “read ahead” document contained a much more comprehensive assessment of several variables that could be used to assess strength and competitiveness. Consider refining the metrics used in this evaluation.
- The role of EFRCs in the materials strategy was not made clear to the committee.
- Early career staff are still not strongly engaged in the materials strategy development. An ongoing effort will be required to involve the staff in the implementation plan.
- Early career staff were uniformly complimentary of mentors and managers. All were equally enthusiastic about the people they have worked with and the intellectually stimulating environment, as well as the resources available to them to conduct science.
- The same early career staff were also uniformly critical of institutional barriers to conducting their work.
- Some of the better, more creative work was unfunded.

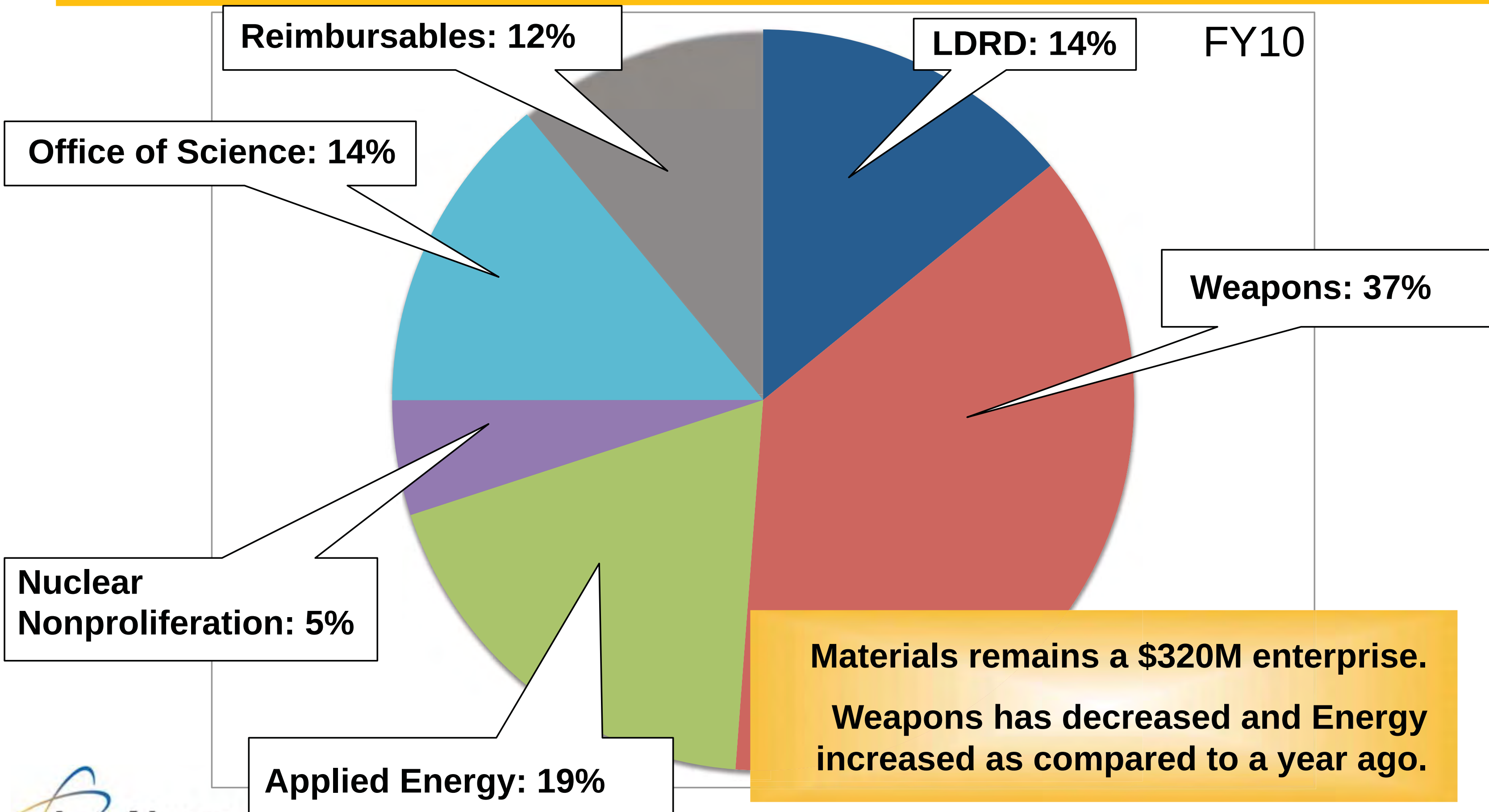
Presentation outline

- **2011 Topics**
- **Response to 2010 recommendations**
- **Trends in metrics**
  - Budgets
  - People
  - Milestones & publications
- **Technical highlights**

The Materials Capability spans the Laboratory, involving ~16 divisions in 7 directorates



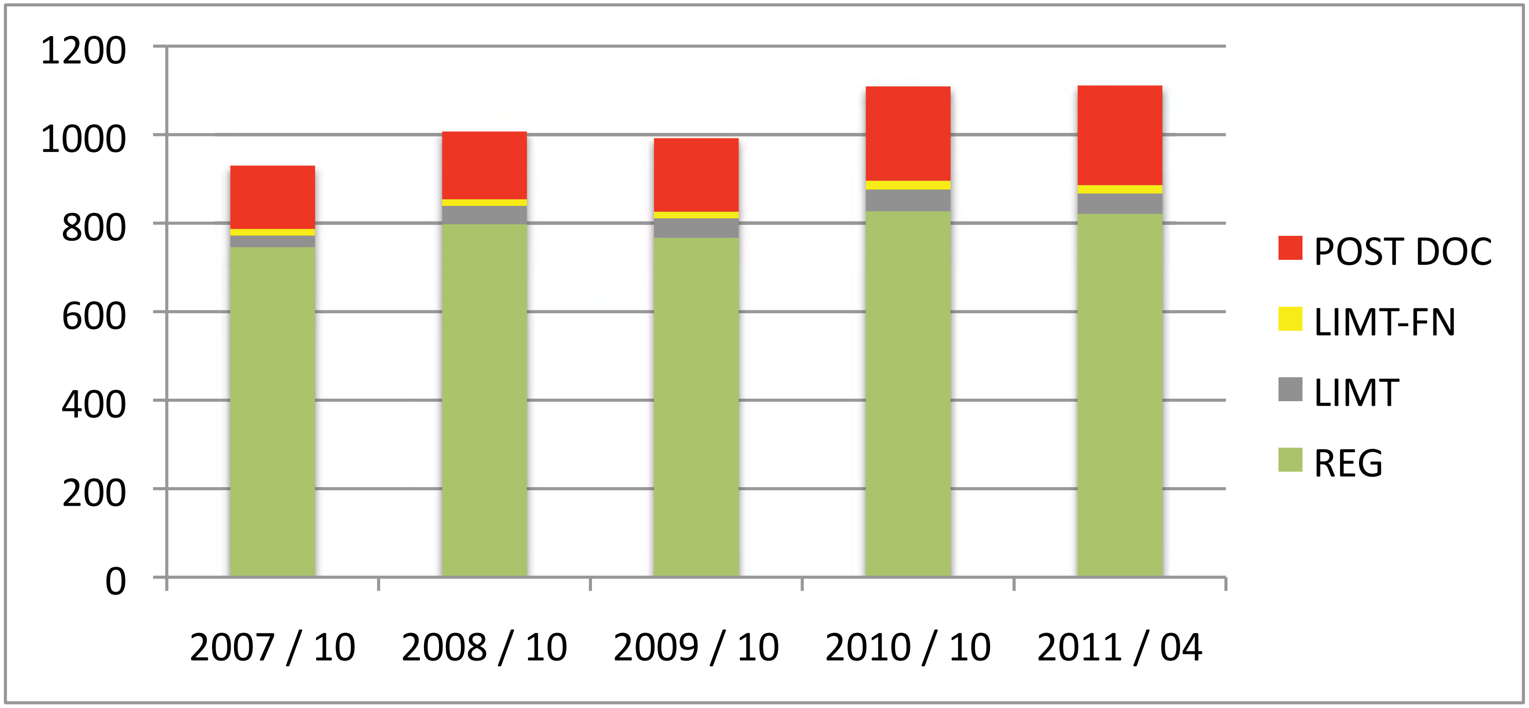
Materials R&D at LANL serves the full suite of Laboratory missions



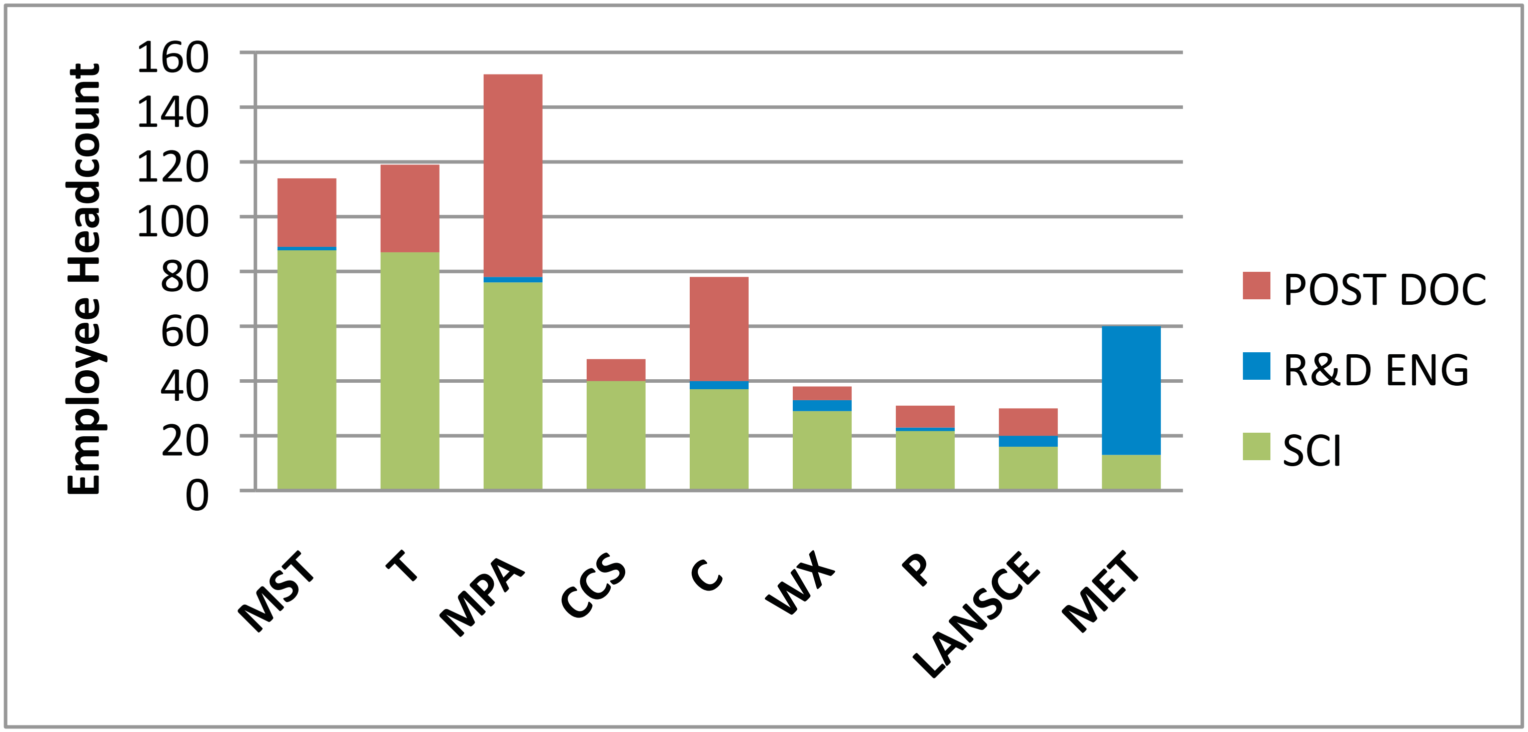
Materials remains a $320M enterprise.

Weapons has decreased and Energy increased as compared to a year ago.

Materials includes approximately 1100 technical professionals

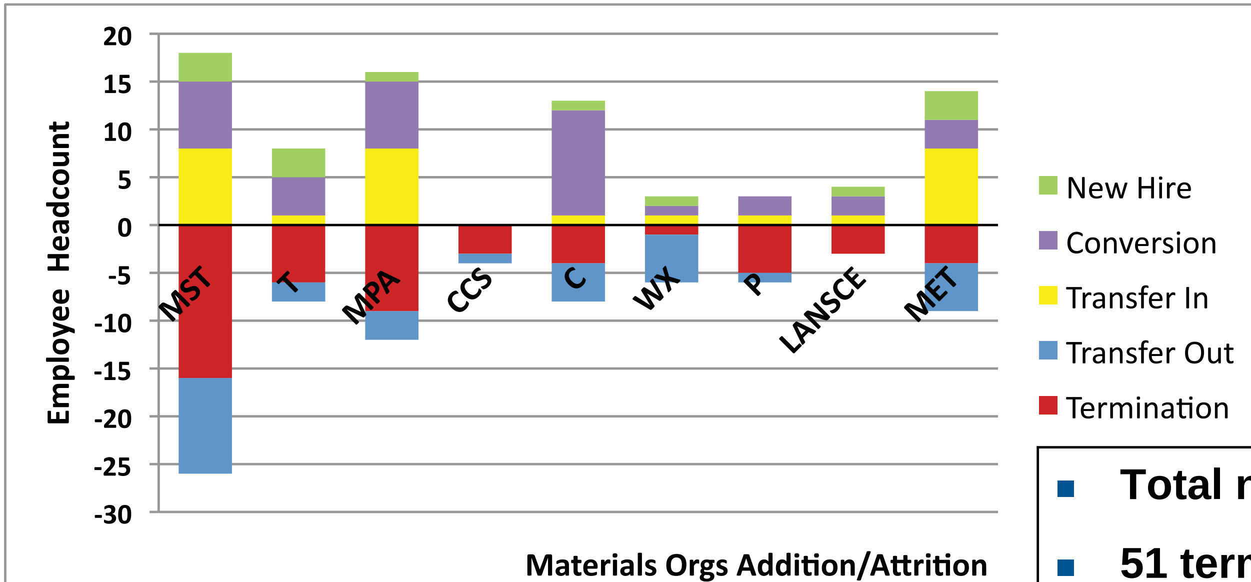


The 2011 demographics of postdocs to staff ratios vary widely across materials organizations



Note: Includes only those portions of each division that are materials-centric.

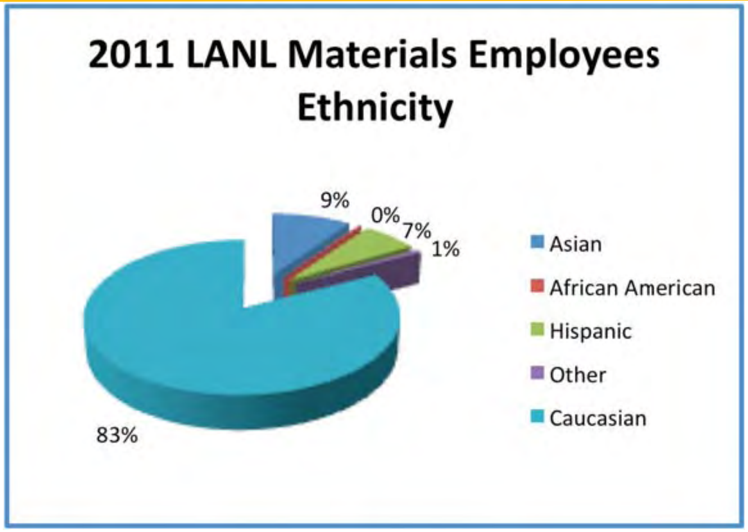
Additions to our organizations occur mainly through conversion of postdocs and internal transfers



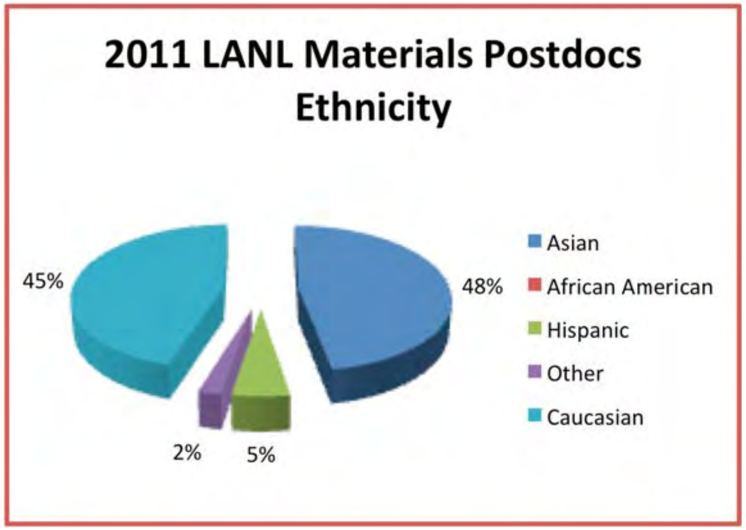
- Total net change -3
- 51 terminations
- 31 transfers out
- 29 transfers in
- 37 conversions
- 13 external hires

- 2+ year trend (10/1/09 – 4/25/11)
- Turnover of ~4 - 7% per year

We need to use our postdoc program to increase our ethnic diversity



Population consists of Scientists, R&D Engineers, and Tech Program Managers. Materials = 356.

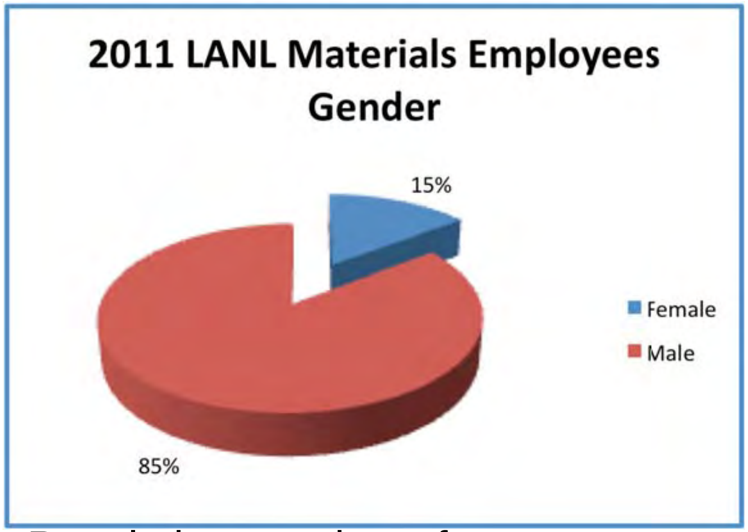


Materials Postdocs = 188.

Are we keeping a good ethnic mix in conversion? An example:

- **Since 2008, 41 postdocs have been employed in MST-8**
- **21 were from Asia (50%)**
- **7 of the 41 were converted to TSMs (15%)**
- **Of those converted, 2 were from Asia (17%)**

We struggle with gender diversity, even in the postdoc program



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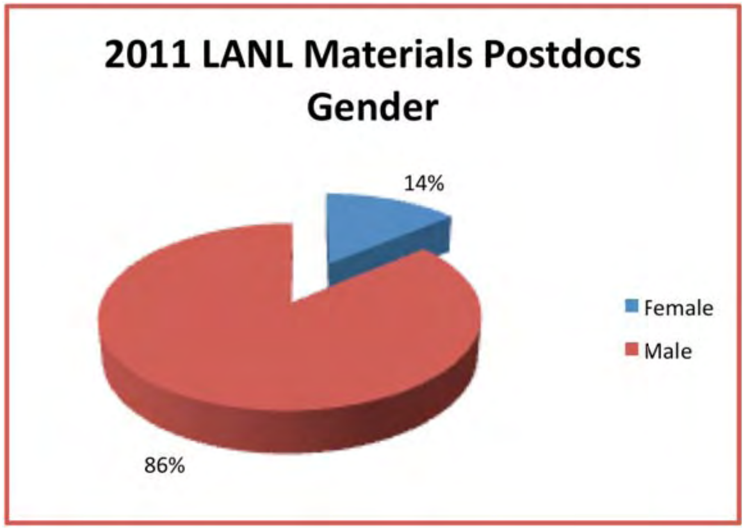
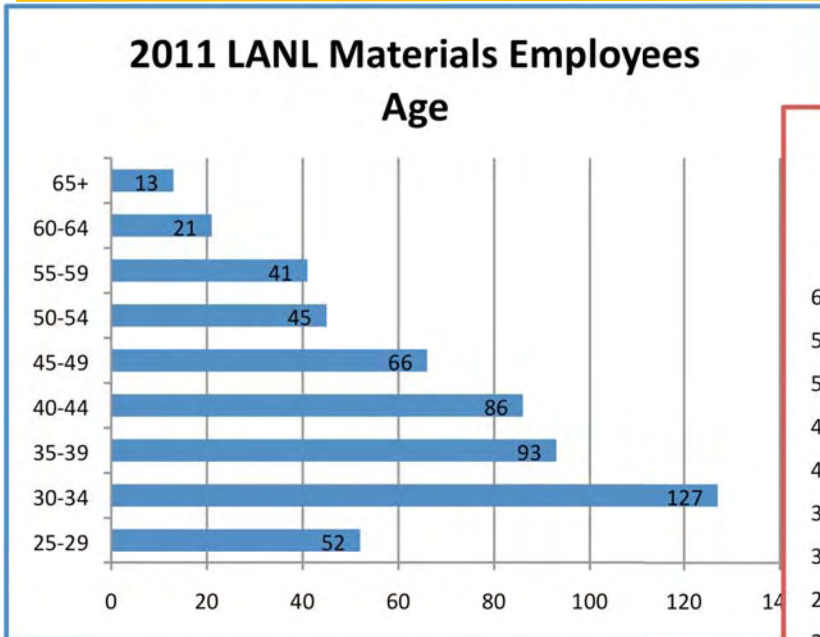


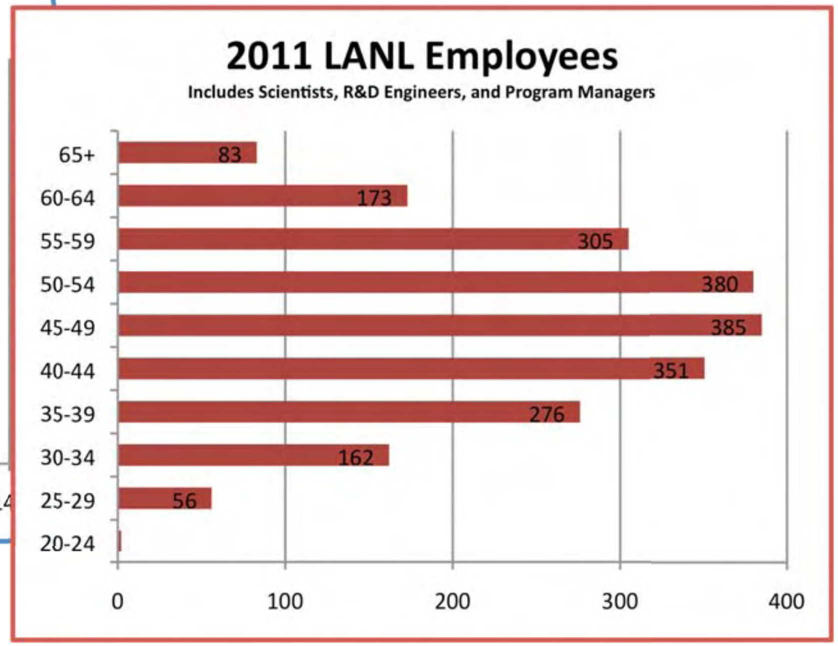
Materials Postdocs = 188.

<u>What is the national availability?</u> *- NSF Report dated 2011 (11-309)*

- **Doctorates in the physical sciences awarded to women in the US have grown from 20% to 30% over the past 20 years.**
- **Women full professors in S&E has grown from 8% to 20% over 20 years.**

Average age for materials professionals is 42, as compared to 48 for the overall LANL population





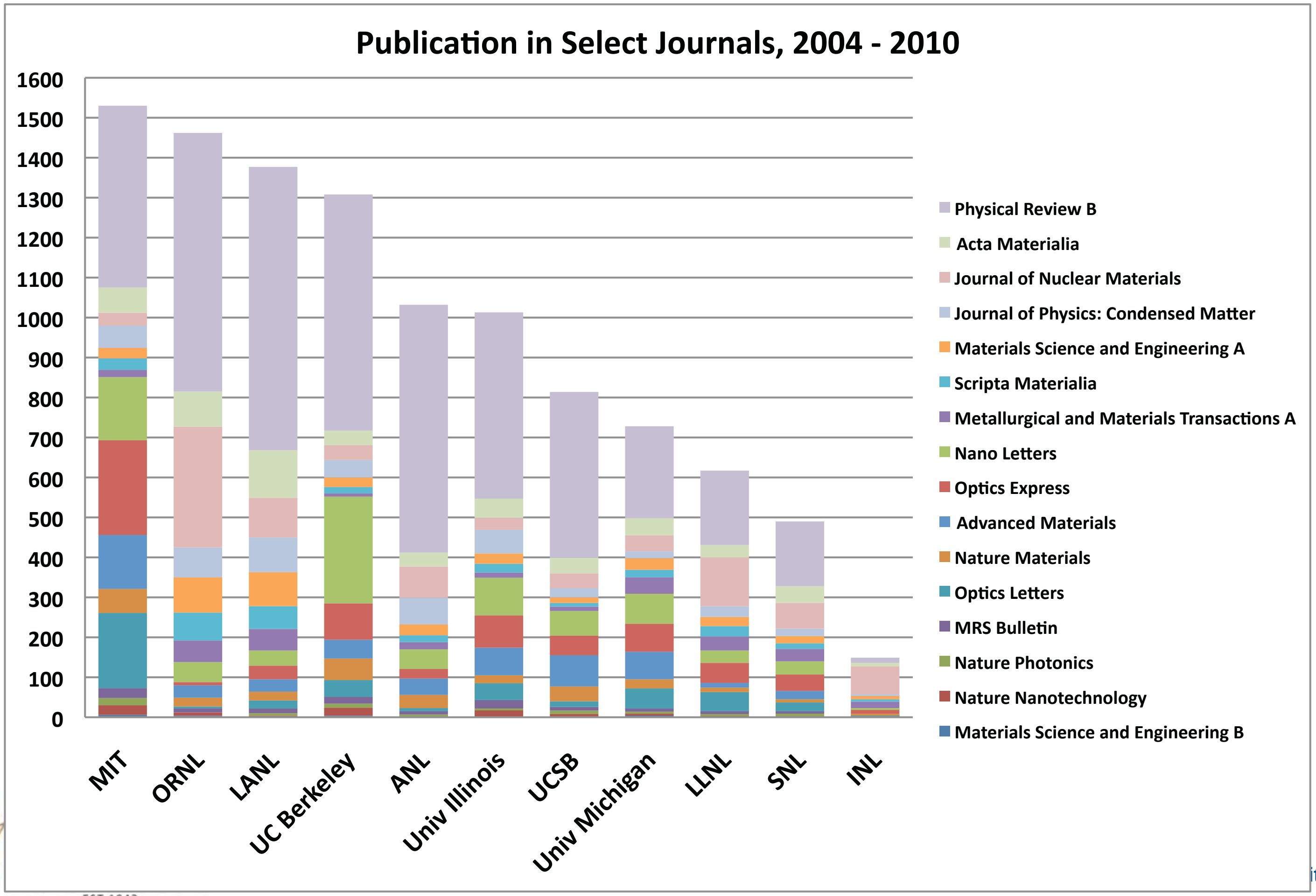
Building the future will rely upon developing and retaining our early career staff!

The materials community has a significant role in weapons program L2 milestones (FY10)

- **305 L2 milestones in DOE complex (many shared with other sites)**
- **83 L2 milestones have materials science input/involvement**

3125	Execute first experiment in Barolo series.
3134	Complete first Pu experiments on the refurbished Z
3341	1.2 - Execute B61 LEP Phase 6.2 study in FY2010.
3399	A13.01-Pu Sustainment-Manufacture 5 Pits that meet final assembly requirements
3404	A13.06-Pu Sustainment-Near Net Shape Die Casting Demonstration
3410	3.3 - Successfully perform high-priority DP mission- related science experiments.
3440	Demonstrate NIF neutron imaging system performance on OMEGA
3467	Provide Pu aging data to support the pit lifetime assessment update in the annual assessment
3513	Phase-aware strength data to support multi-phase strength and damage model development
3514	Provide assessment of multiphase metal EOS against dynamic and static data

LANL is one of the most prolific organizations in the materials literature



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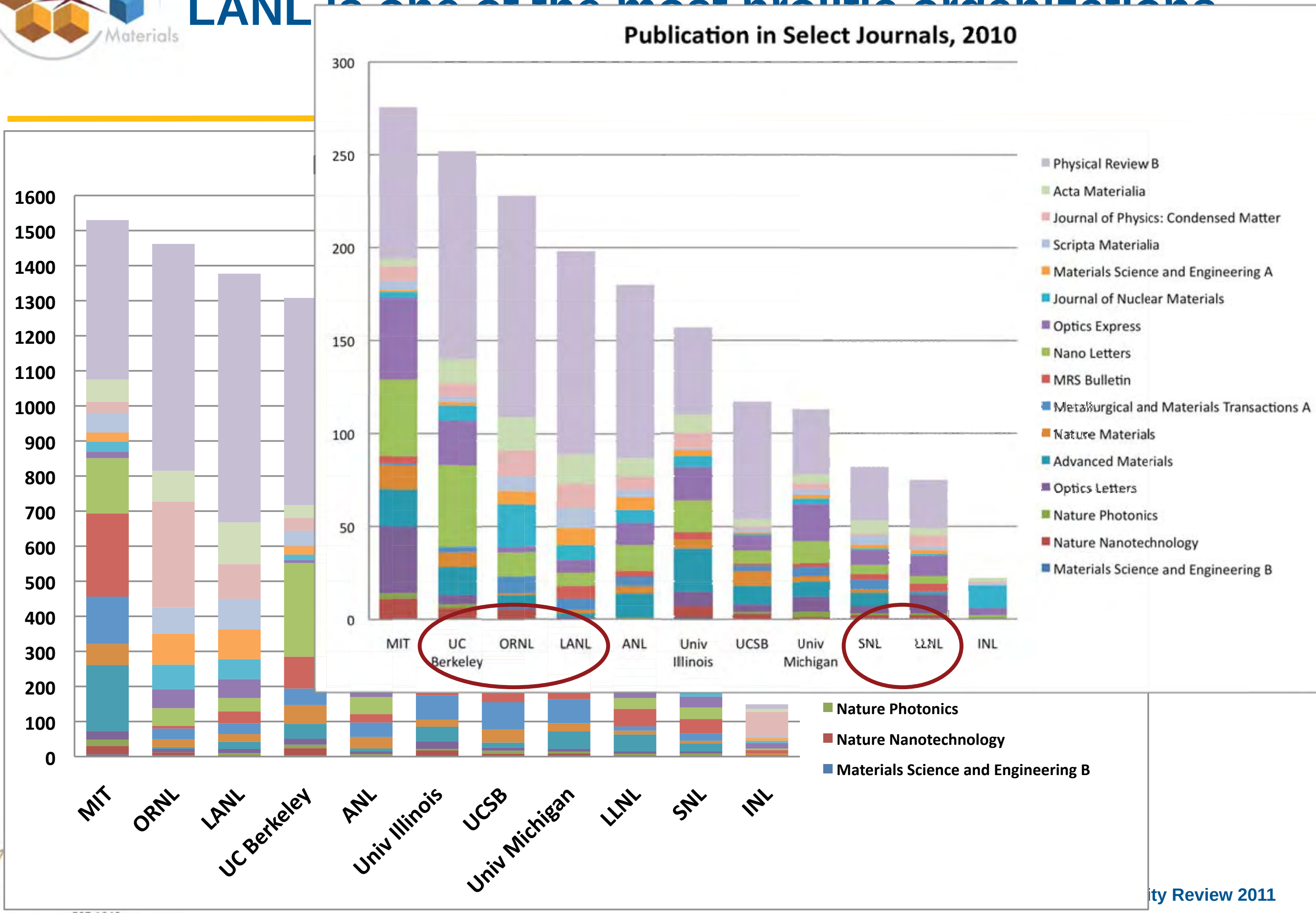
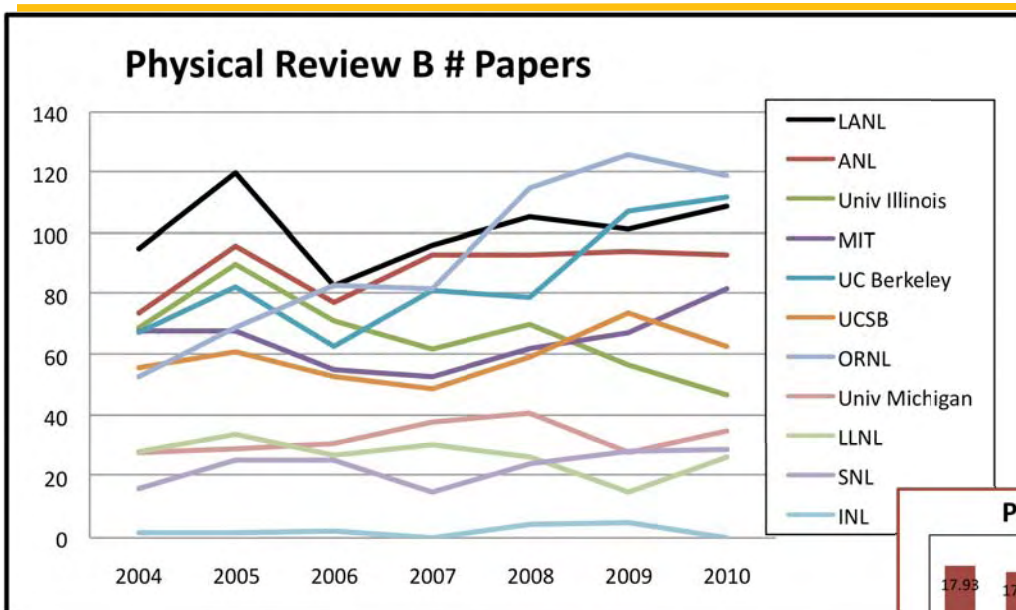
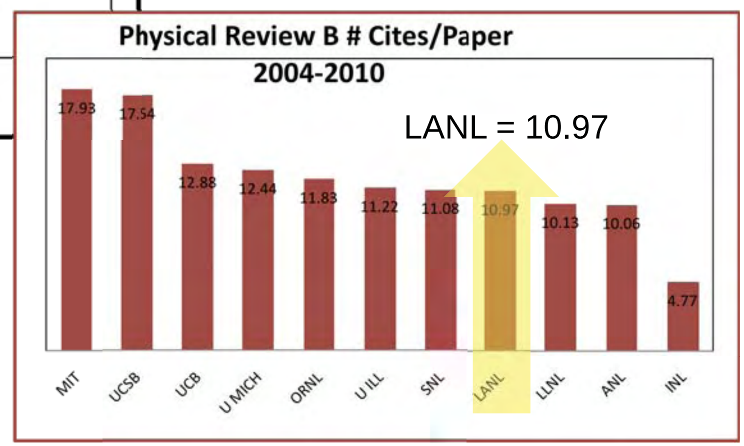


Trends in publications in *Phys Rev B*

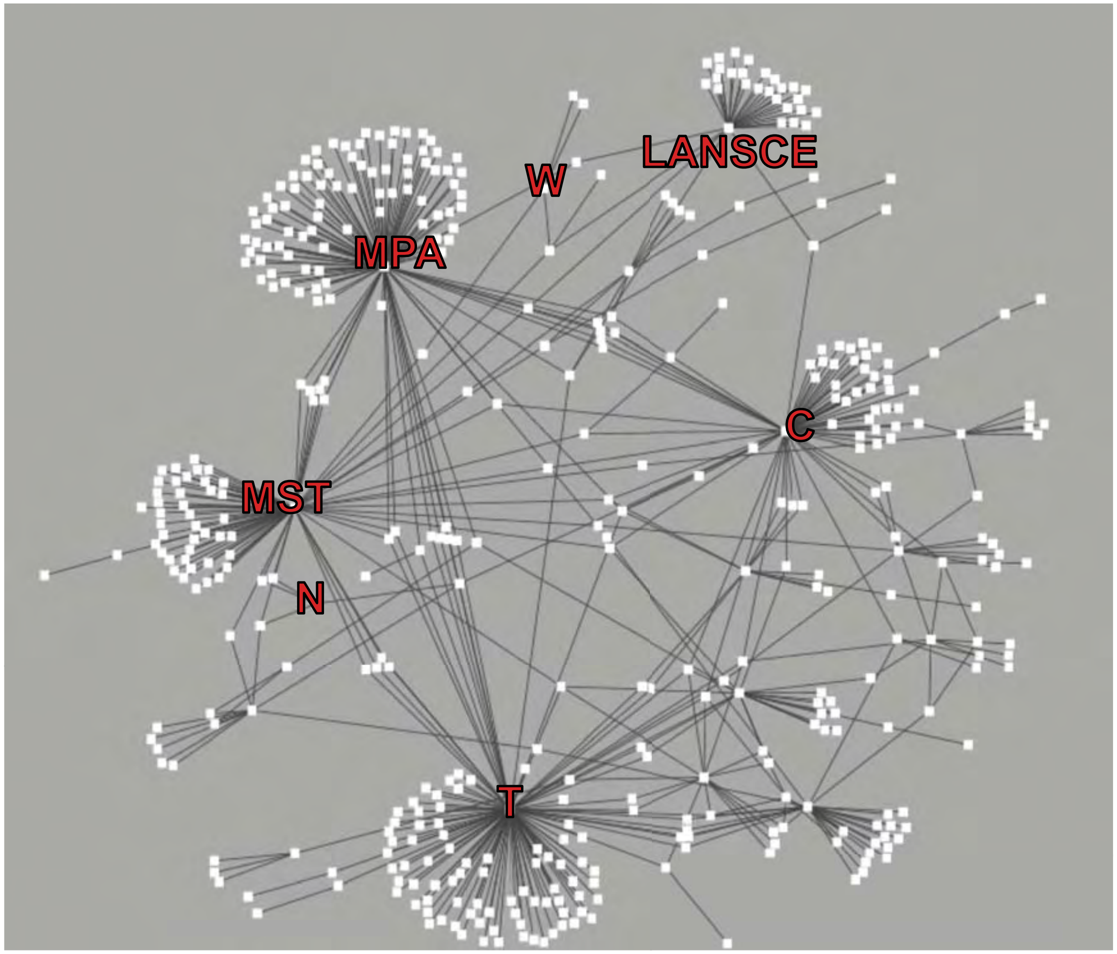




Phys Rev B average cites/paper = 10.03

New mapping techniques depict our interdisciplinary collaboration

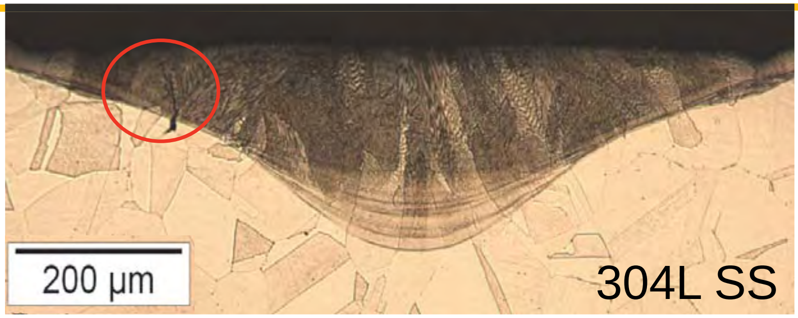
- LANL Research Library database (ISI, Inspec, BIOSIS, Engineering Index)
- 745 employees
- 902 publications in 2010
- 2568 LANL authors
- 16 divisions
- Points are article titles and divisions

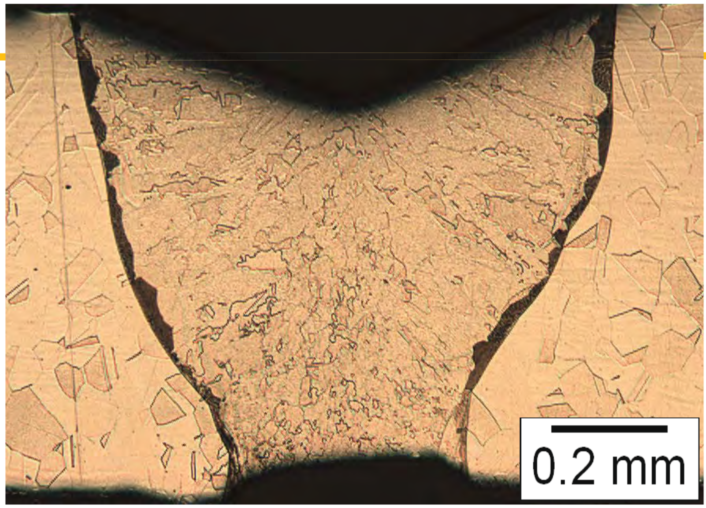


Presentation outline

- 2011 Topics
- Response to 2010 recommendations
- Trends in metrics
- **Technical highlights**

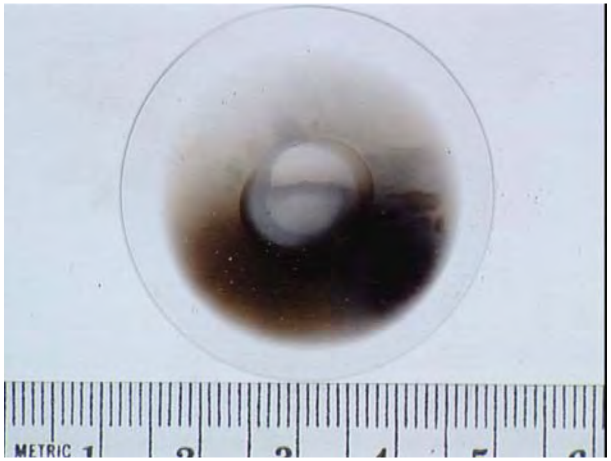
Controlling functionality through making: Nondestructive laser gas sampling

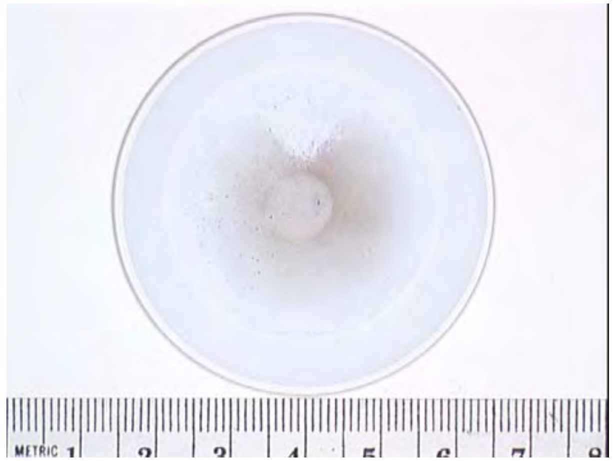




Issue 1: Solidification cracking eliminated by Cr additions

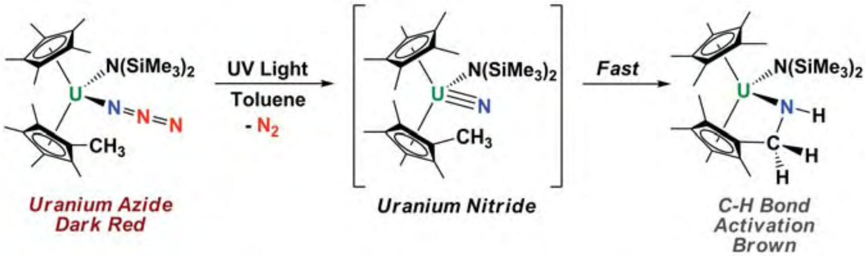
Issue 2: Window "fogging" eliminated by adjusting parameters to limit evaporation





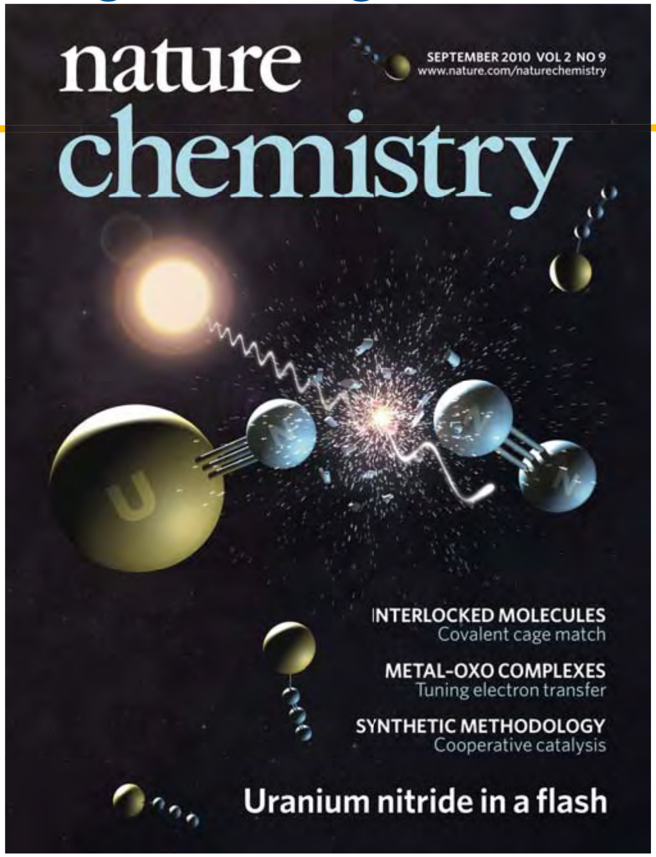
T. Lienert et al

Controlling functionality through making: Uranium azide photolysis



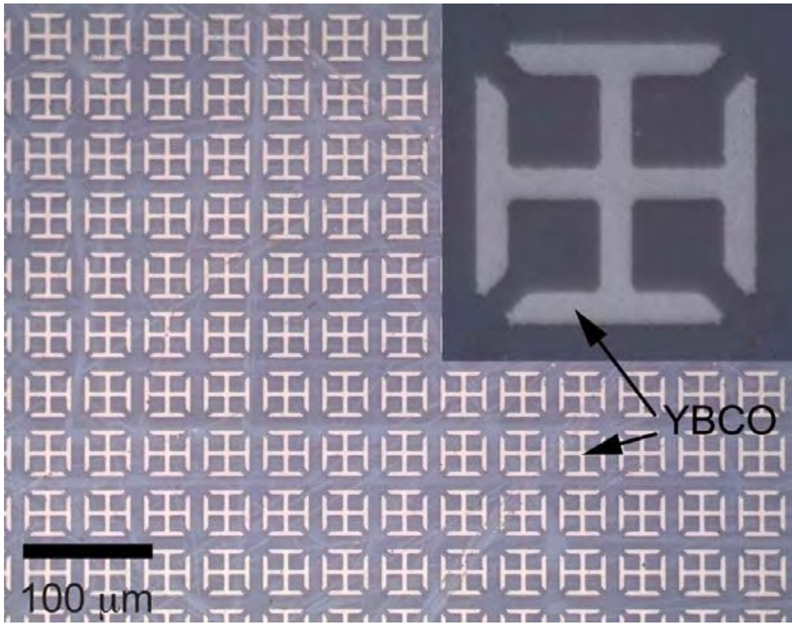
- The first ever evidence for the formation of a uranium nitrido complex that contains a terminal U≡N bond.
- Photochemistry is shown to be a powerful synthetic tool for performing molecular transformations in actinide chemistry.
- The U≡N complex is highly reactive, able to attack strong C-H bonds to form new N-H and N-C bonds.
- The U≡N unit is not inert and can undergo reactions with strongly-bonded molecules.

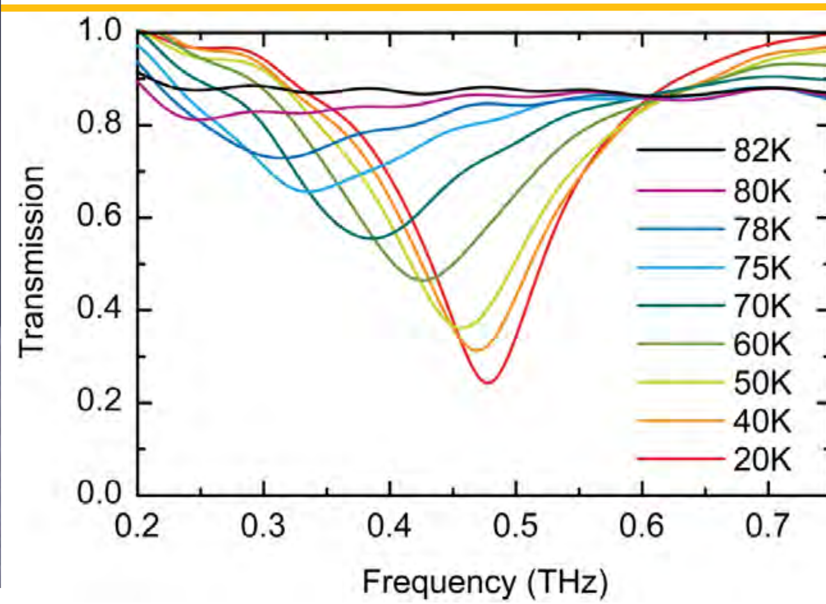
"Uranium azide photolysis results in C–H bond activation and provides evidence for a terminal uranium nitride," Robert K. Thomson, Thibault Cantat, Brian L. Scott, David E. Morris, Enrique R. Batista & Jaqueline L. Kiplinger Nature Chemistry 2, 9, 705 (2010)



Depiction of a uranium azide complex ($U\text{-}N^3$) interacting with light to lose nitrogen and produce a terminal uranium nitride complex (U≡N).

Controlling functionality through making and measuring: Superconducting terahertz metamaterials





- **Fabricated terahertz metamaterials from YBCO high-temperature superconducting films (50-200 nm) thick**
- **Tuned the resonance of YBCO metamaterials by temperature, both the resonance strength and frequency**
- **Described the tuning mechanism by relating the real and imaginary conductivity to the resistance and inductance of the split-ring resonators**

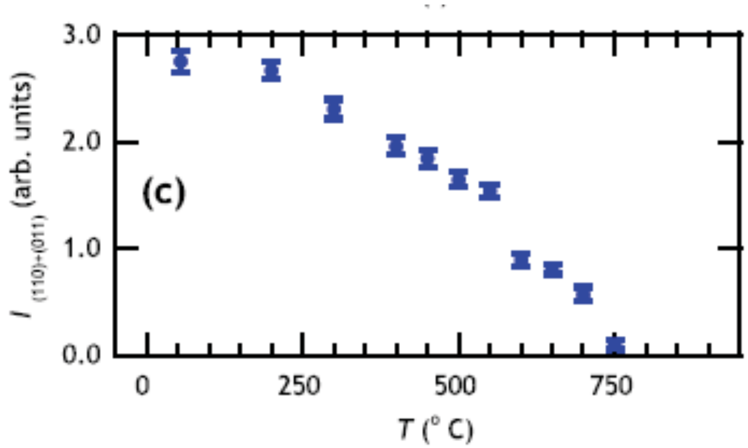
*H.-T. Chen et al., PRL, **105**, 247402 (2010)*

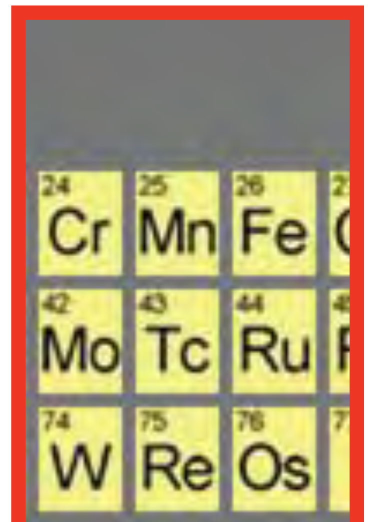
Controlling functionality through measuring: New Tc perovskite $SrTcO_3$

- **Technetium is an unexplored radioactive kin of molybdenum and ruthenium**
- **$SrTcO_3$ has the highest magnetic ordering temperature in any compound without 3d transition elements.**

PHYSICAL REVIEW LETTERS

Rodriguez et al, PRL, 106, 67201(2011), Feb 11 2011



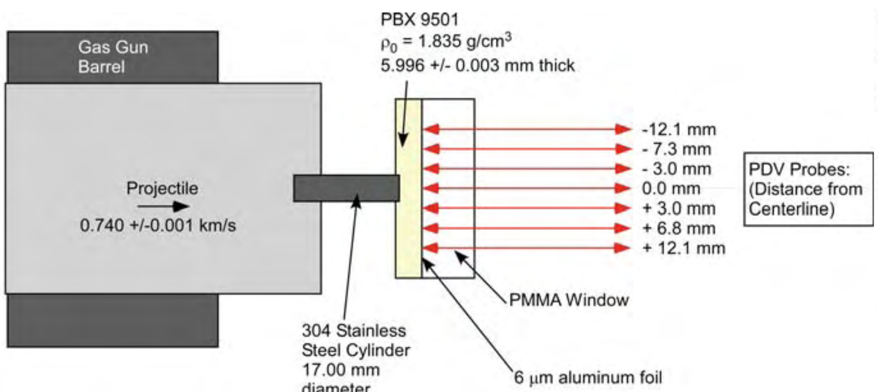


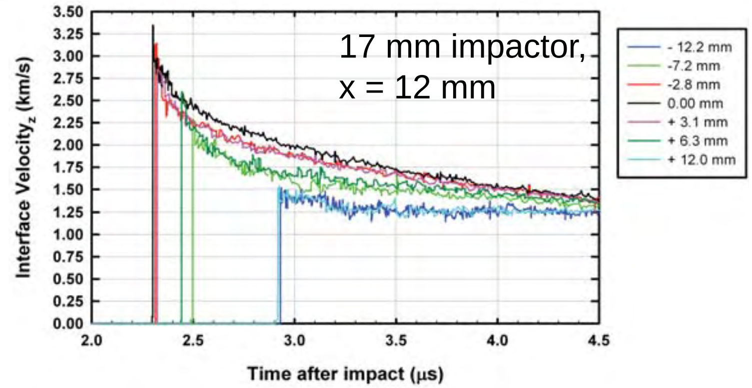
Results: $SrTcO_3$ adopts a distorted perovskite structure with antiferromagnetic ordering. The magnetic order persists up to an extraordinarily high Neel point, approximately 1000K.

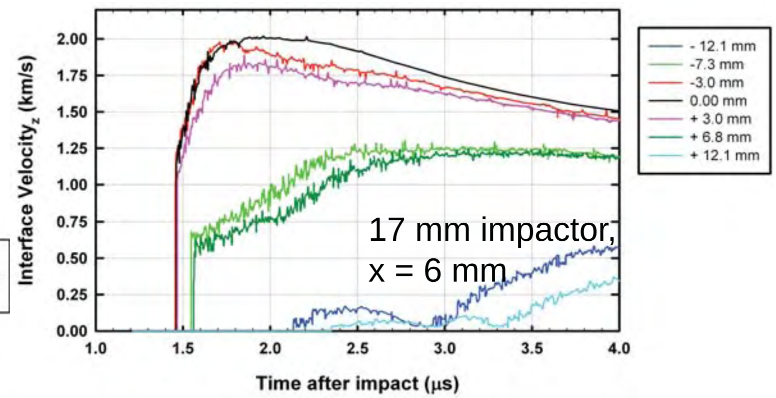
Controlling functionality through measuring: The role of shear in the initiation of explosives

- Well-defined combined shock-shear loading conditions
- PBX 9501 explosive
- **Observing little effect of shear, i.e. initiation is dominated by pressure/impulse**
- Data will establish key factors in bullet and fragment M Hazard

Gustavsen, Dattelbaum, Johnson

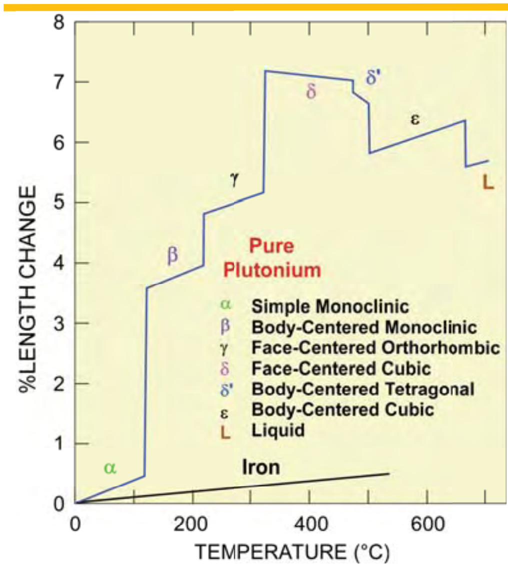




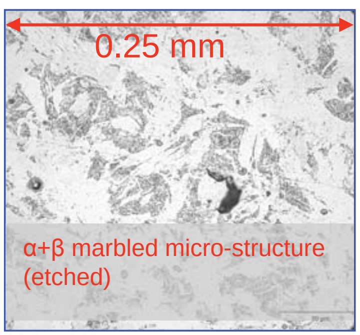


Multi-point PDV diagnostics- shock wave profiles at defined run distance

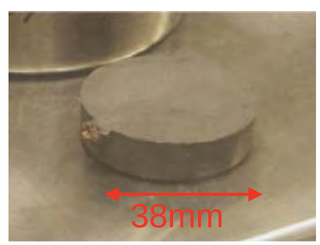
Controlling functionality through modeling and making: Alpha phase plutonium



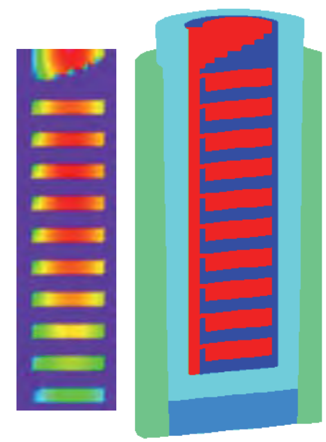
- Unalloyed Pu castings undergo >24% volume reduction from liquid to RT solid.
- Metallurgical and casting "tricks" get you close to α-Pu (19.86g/cc)
  - Ga microalloying (~500 ppm pins cracks)
  - Pre-heat
  - Puck casting
  - Thermal cycling



19.62 g/cc



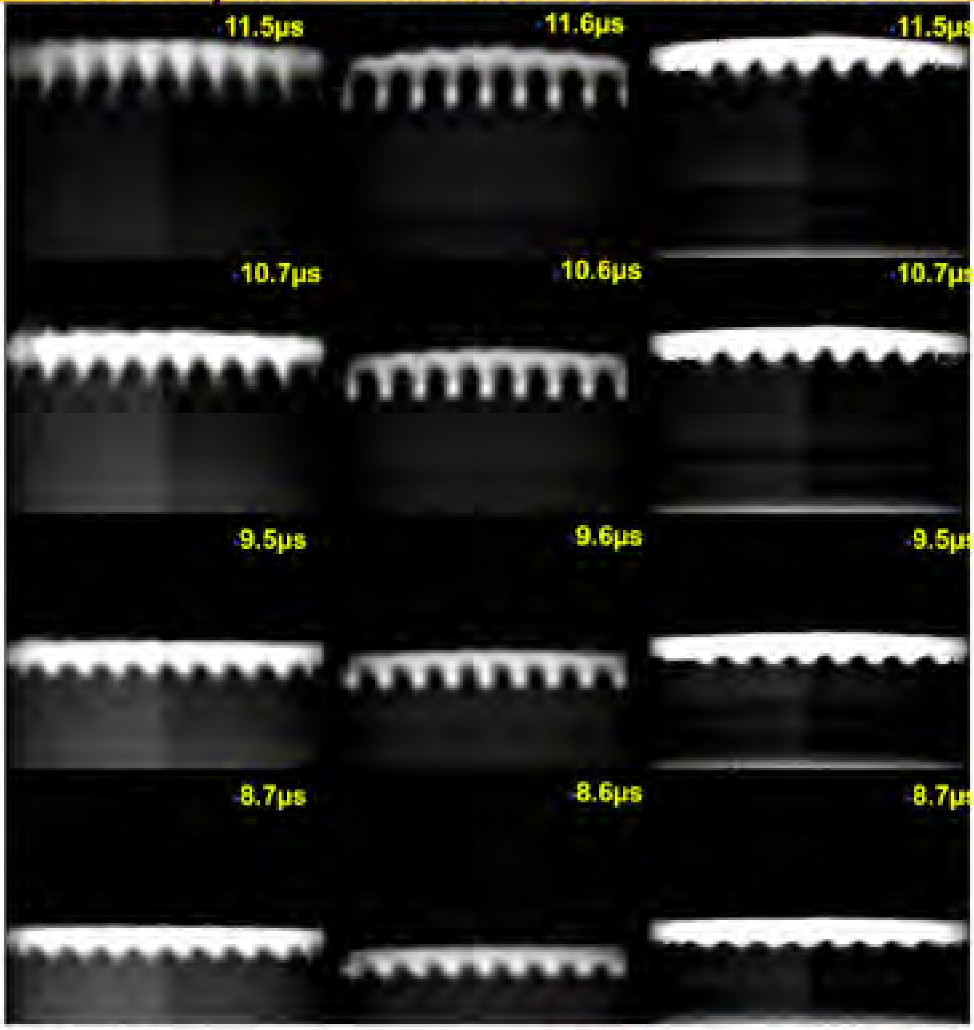
Puck Casting Models
Simulation of casting showing time to solidify



Korzekwa, Freibert

Controlling functionality through measuring and modeling: Strength-inhibited R-T growth experiments validate strength models at high strains (>100%) and strain rates (10^5-10^7 s^{-1})

- **Cu samples tested with identical drive conditions and initial perturbation: (3.0 mm vacuum gap, λ=2.0 mm, A_0=55 μm)**
  - Annealed Cu: 60 μm average grain size
  - Cold-worked Cu: 30% strain, cross-rolled, deformation twins
  - Single crystal Cu: loading direction along 001 and 101 orientations
- **Perturbation growth rate and shape depend strongly on microstructure.**
  - Improved physics-based strength models must capture 'sub-grid' material behavior to accurately predict bulk hydrodynamic behavior.

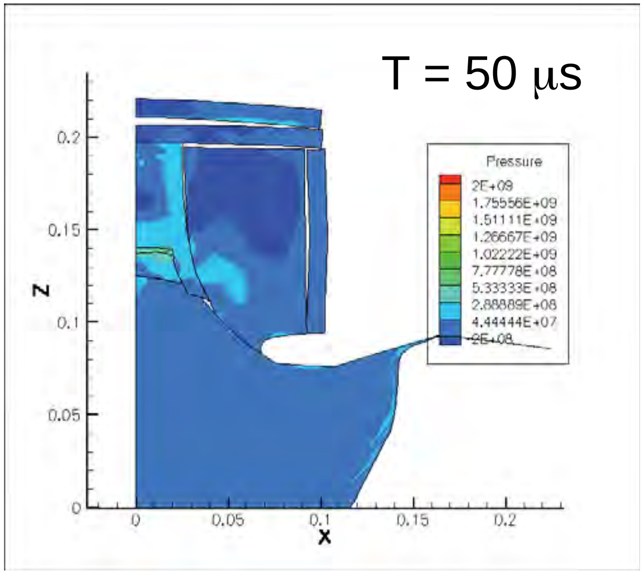


Cold-rolled Cu Micrograph

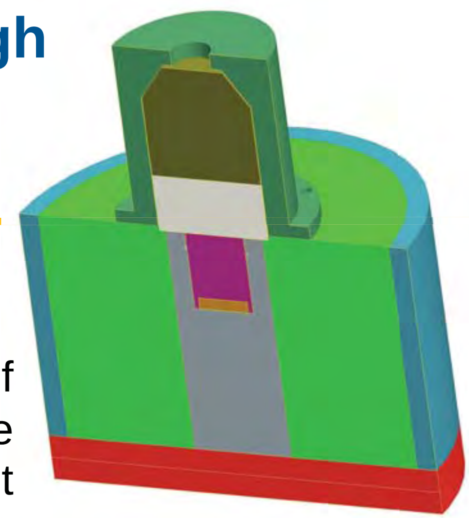
R. Olson et al

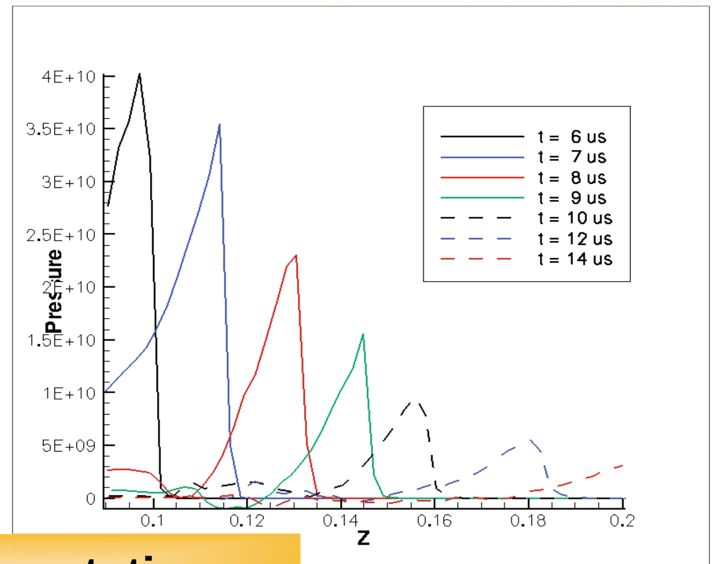
Controlling functionality through measuring and modeling: Be EOS and plasticity

Addessio, Bronkhorst



Final design of the HE driven Be experiment





Simulations of the HE driven experiment:
Above - contours of pressure profiles
Right - pressure distributions along the axis

The computational representations accurately reproduce experiment

Your feedback on the performance of the Materials Capability is requested

- **Scientific Leadership as Evidenced by:**
  - Leadership in an international technical community
  - Publication of highly cited research results
  - Flux of innovative ideas and proposals
  - Hiring and training of next generation's leaders
- **Programmatic Impact**
- **Sustaining a *Cross-Laboratory* Materials Community**
- **Direction for the Future**
  - Progress on implementing the Materials Strategy
  - LANSCE and the path to MaRIE
  - Recapitalization and facilities revitalization

Materials for the Future:
Towards an implementation plan for the Materials Strategy

A.J. Taylor (MPA-DO)

We are embarking on a new era in materials science, where we transition from observing and exploiting the properties of materials to a science-based capability to create materials with properties optimized for specific functions. This concept of “controlled functionality” is central to LANL’s Materials Strategy. This vision of intentional control of functionality will be realized through discovery and application of materials synthesis, fabrication, and processing, materials characterization, and theory modeling and simulation, across the three theme areas of Emergent Phenomena, Defects and Interfaces, and Extreme Environments.

In this presentation I will describe our progress towards developing an implementation plan for the Materials Strategy. This process, which has engaged more than 100 scientists spanning four of the Laboratory’s directorates, has resulted in the identification of six areas of leadership for LANL’s Materials for the Future, identifying ~$25M thrusts within each of these areas. Key science questions, along with a ten-year programmatic and capability roadmap, were developed for each of the thrusts. Cross-cutting strategic opportunities were then identified and are being incorporated into a draft implementation plan for the Materials Strategy.

Materials for the Future:
Towards an Implementation Plan for the Materials Strategy

Materials Capability Review 2011

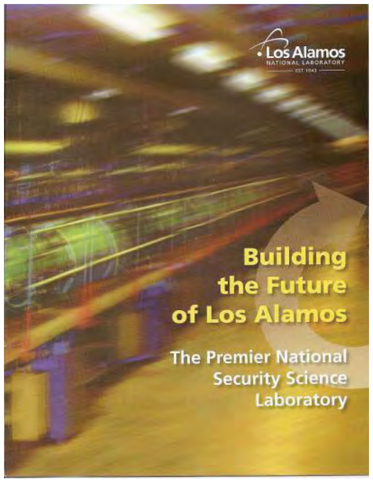
Toni Taylor
Division Leader
Materials Physics and Applications

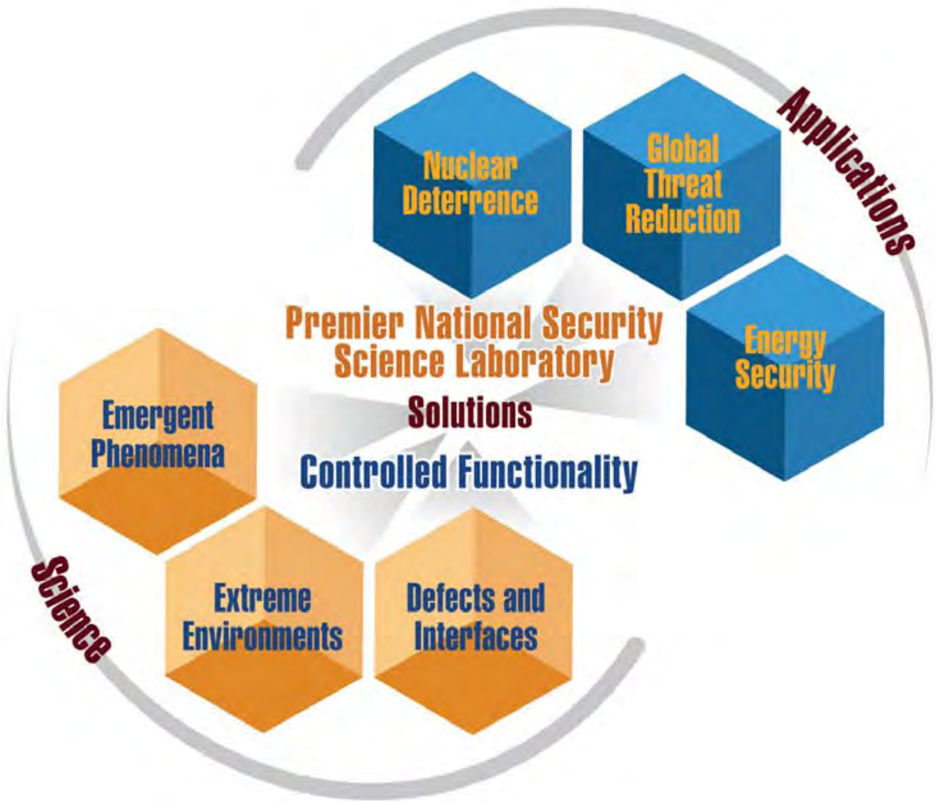
Operated by Los Alamos National Security, LLC for NNSA

Outline

- Overview of Materials Strategy
- Process to define and develop a vision for the 'Areas of Leadership'— 'Deep Dives'
- Outcome of Deep Dives
- Draft implementation plan
- Path forward

The Materials Strategy advances our vision to develop materials with 'controlled functionality' to provide solutions enabling LANL's missions





Within the 3 science themes, the S&T that comprises LANL's Materials Capability needs to be identified and current status defined

Areas of leadership for LANL Materials (preliminary)

Nuclear Deterrence

- Actinides
- Materials dynamics
- Energetic materials

Global Security

- Materials attribution
- Materials and diagnostics enabling sensors/detectors

Energy Security

- Fuels, targets and structural materials for fission/fusion
- Next generation materials for energy utilization

Crosscuts: User Facilities, Theory, Fabrication, Characterization

- Multifunctional and adaptive materials
- Correlated electron materials

Emergent Phenomena

- Creating and diagnosing extreme conditions
- Materials under radiation extremes

Extreme Environments

- Designer materials optimized for specific needs
- Integrated nanomaterials

Defects & Interfaces

Correlated Electron Materials

Input by: Thompson/July, 2010

Key Topics:

- New physics through new materials – emergent phenomena
- Pu and Pu-based materials
- Unconventional superconductivity/materials
- Quantum criticality/quantum states
- Coupled spin-charge-orbit-lattice degrees of freedom, eg. multiferroicity, geometric frustration
- Extreme environments/tuning parameters/diagnostics (low T, high P, high H)

Key personnel/expertise needs:

- Mid-career, internationally-recognized intellectual leadership, esp. experimental
- Spin spectroscopies (solid-state NMR, neutrons)
- Crystal growth of new materials
- Very low-temperature scanning spectroscopies
- Internationally competitive postdocs
- Spin-resolved charge spectroscopies (ARPES,STM)
- Ultrafast spectroscopies
- MBE film growth

What is missing in the alignment of program pull" and science "push"?

- Institutional flexibility to be responsive to new discoveries
- Coordinated, appreciated continuum from basic to applied
- Appreciation that science is 'fragile'
- Accelerating/amplifying LDRD -> BES transition at a level to sustain capability
- Better connection/coordination with MaRIE

Key facility and equipment needs:

- Modernized facilities
- Co-location of basic, applied, experiment and theory
- Facility for handling small quantities of transuranics without TA55 'overhead'
- Helium liquefaction
- Inelastic scattering at Lujan

Key operational barriers:

- Need for a CNLS-/CMS-like facilitating organization
- Need for a sense of community/shared fate within LANL, including understanding trade-offs, agility requirements
- Turf-protection limiting cooperation/leverage

Competition

- Max Planck Institute for Chemical Physics of Solids (Dresden) – if LANL is #1 or #2, then this is the other
- CRNS/CEA, Grenoble
- China!
- Japan Universities, esp Kyoto and Tokyo, and JAEA—transuranic research
- Neutrons: SNS, NIST, ILL, PSI, Berlin
- To some extent DOE labs, eg. ORNL, BNL, Ames
- To some extent US and European universities, esp. in theory

Issues identified through the process of developing the Materials Strategy

Program/capability/science interface

- Much improved coordination required between short term deliverables required by programs and long term capability needs
- Institutional flexibility to be responsive to new discoveries
- In areas of strategic importance, the support of a coordinated continuum from basic to applied R&D
- Institutional appreciation that science is ‘fragile’

Facilities/operations

- Modernized facilities that are inherently HPI
- Co-location of basic, applied, experiment and theory
- Ability to handle small quantities of transuranics without TA55 ‘overhead’
- Functional models for operating mid-scale user facilities like the Electron Microscope Laboratory
- Barriers to R&D induced by inefficient/risk adverse safety and security processes

Personnel/outreach

- Recruitment and retainment of personnel in key technical areas
- The ability to attract mid-career leaders to the Lab
- Connecting people to new programs as funding and science evolves
- Lack of institutional prowess in contracting/partnering with industry & national labs
- Access to facilities by visitors, esp. foreign nationals

2nd iteration: The Areas of Leadership span the 3 themes of the Materials Strategy

Extreme Environments

- Materials for nuclear energy/extreme radiation environments
- Materials dynamics
- Energetic materials

Defect and Interfaces

- Materials & diagnostics enabling sensors, detectors, & attribution
- Actinides
- Integrated nanomaterials

Emergent Phenomena

- Correlated electron materials
- Multifunctional and adaptive materials
- Materials for energy utilization

Crosscuts: Making, Measuring and Modeling

Developing an Implementation Plan: What actions are essential to our vision for Materials S&T?

- **What capabilities are central to our areas of leadership, and how must these evolve over the next decade?**
- **Who are our competitors, and/or our collaborators?**
- **How can we leverage current programmatic efforts and develop new programs that enable our vision to succeed?**
- **What infrastructure is critical to our success, and how must this evolve?**

Deep Dives:

Engage LANL Materials community in developing vision in areas of leadership

Deep dives implemented via a series of half-day workshops involving scientists and leaders

- Actinides / Correlated electron materials (EP/ME)
- Integrated nanomaterials / Materials for energy utilization (EP/DI)
- Materials dynamics / Energetic materials (ME/DI)
- Materials & diagnostics enabling sensors, detectors, & attribution / Multifunctional and adaptive materials (EP/DI)
- Materials for nuclear energy (DI/ME)

Deliberate mixing across Materials theme areas

Participation by invitation, as suggested by Materials division leaders

Engaged >100 scientists across four Laboratory directorates

Deep Dives: A more forward looking perspective engaging key scientists across the Lab

Nonlinear metamaterials

Nanoclusters for sensing

BES Core

Emergent Functional Materials

EFRC CASP

Complex oxide Interfaces

Superconductivity by design

Create a vision and roadmap for each area of leadership

- Materials R&D is currently ~$300M at LANL – Can we identify ~12, $25M "big picture" concepts supporting the areas of leadership?
- Roadmap: identifying customers, facilities, equipment, staffing needs

Example: Actinide & correlated electron materials: Key science questions

- **How do correlations determine functionality in actinides?**
- **How do we develop predictive tools to allow materials design for functionality (electronic structure → observables)**
  - Can we model and predict the multi-component phase stability and its impact on performance?
  - Can we understand and predict interactions with the environment?
- **What is the interplay between structure and correlations, and what are the consequences therein?**
  - How do we understand the Structure/Property relationship across Pu phase space (pressure, temperature, time, composition, strain rate)?
  - What are the relevant length and time scales for plutonium science?
  - How do we model/predict and develop new theory to describe the unique properties within plutonium?
- **What is the effect of defects and interfaces (includes surfaces) at the nano to microscale?**

Actinide & correlated electron materials end states with thrusts

- **"Understanding and controlling emergent electronic states"**
  - Tuning of correlations in extreme environments
  - Functionality from coupled degrees of freedom
  - Functionality from quantum criticality
  - Superconductivity
- **"Actinide materials science center of excellence"**
  - Structure/Property relationships from ambient to extreme conditions
  - Plutonium properties across relevant length scales
- **"Predicting and controlling plutonium aging and lifetime"**
  - Validity of "Acceleration" in predicting lifetime, including the identification of fundamental control variables (Dose, Dose Rate, Temperature)
  - Development of a robust QMU framework for actinide materials
    - Intrinsic Variability – Stochastic processes, materials variation, etc.
    - Role of Pedigree and Well-characterized Materials
- **Cross Cut: Predictive tools to allow materials design for functionality**

Actinide & correlated electron materials: Programmatic road map

End state: Integrated and robust capability utilizing world-leading infrastructure (CMRR, MaRIE, intermediate-scale and public user facilities)

5 yrs: Realization of Plutonium Science & Research Strategy; Hub-scale effort in electron correlation functionality by design

2 yrs: Integrated suite of strategic partners (including for pipeline – e.g., Seaborg); Broadened customer base (e.g., NE, NN)

(Relatively) stable and (uncoordinated) diverse core of programs – Unambiguous international science leadership

Current state

Understanding & controlling emergent electronic states: Additional requirements

Gaps in Current State:
- Predictive electronic structure tools
- Breadth & integration of synthesis capability
- Characterization tools with needed spatial/ temporal/spectral resolution , esp. spin degrees of freedom
- Rare Earth Materials Replacement Opportunity

Requirements at 5 years:
- Crystal growth center
- Time-resolved imaging from infrared to soft x-ray
- Enhanced high magnetic field measurement capability (fast vortex dynamics, imaging)
- Combined capability for high-P, high-H, low-T (coupled to facilities – Lujan, NHMFL, etc.)
- Ultrafast & ultra-small modeling capability for correlated materials

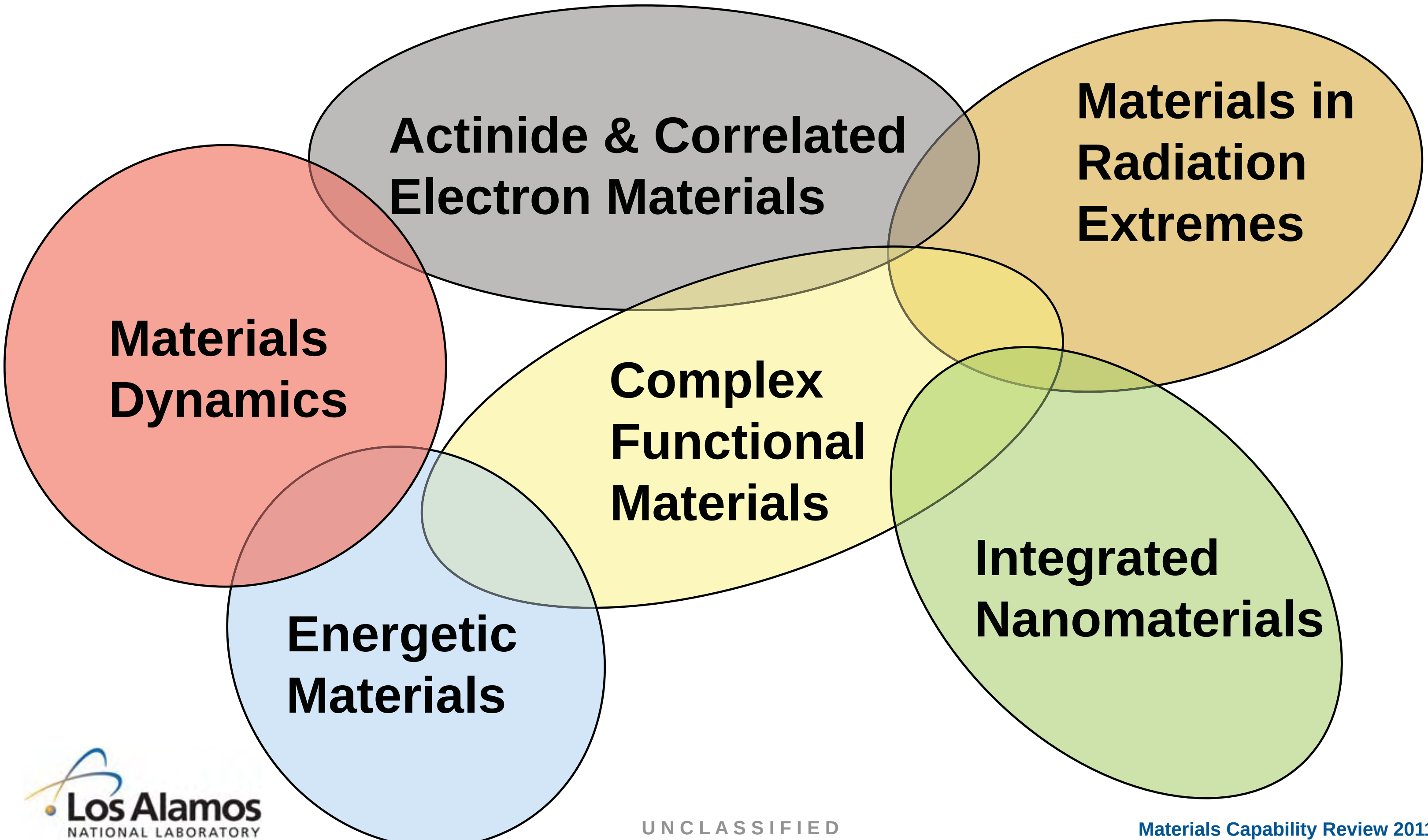
Requirements at 2 years:
- Increase synthesis capability in complex oxides
- High-throughput computing environment
- Facility Integration (Lujan, NHMFL, CINT) bridging to photon sources
- OSTP initiative “computational chemistry & materials science for innovation”

Requirements at 10 years:
- Coupling to MaRIE & M4
- Co-locate experimental facilities crucial for emergent electronic materials research (theory, experimental)

Actinides and correlated electron materials: Strategic implementation opportunities

- Champion and seize "Functionality by Design" Opportunity – OSTP in general, Rare Earths/Fuels for Nuclear Energy in particular (2 Years)
- Establish more robust partnership between materials-centric user facilities –CINT, Lujan, NHMFL and increase footprint (including for actinides) at other public facilities. (2 years)
- Establish capability to translate from fundamental science to technology on a 5-year time frame – Nevada Initiative II (5 years)
- Better integration of experimental and IS&T capabilities in materials design, enabling co-design (5 years)
- Robust implementation of QMU for lifetime acceleration and prediction (10 years)
- Comparable infrastructure and predictive capability (including human resource) for actinides as for non-radioactive materials. (10 years)

Six ‘Areas of Leadership’ span the Materials Pillar



Thrust areas further define the 'Areas of Leadership' for the Materials Pillar

- **Integrated Nanomaterials**
  - Reduced dimensionality materials for control of emergent functionality
  - Center for Nanophotonics
- **Complex Functional Materials**
  - Functional materials for energy conversion, storage and transmission
  - Materials inspired by living systems
  - Multifunctional adaptive materials
- **Materials in Radiation Extremes**
  - Advanced radiation & temperature tolerant structural materials
  - Advanced nuclear fuels & nuclear waste materials
- **Actinides and Correlated Electron Materials**
  - Understanding and controlling emergent electronic states
  - Actinide materials science center of excellence
  - Predicting and controlling plutonium aging and lifetime
- **Materials Dynamics**
  - Linking microstructure to macroscopic behavior under dynamic loading
  - Observation-to-control of dynamic processes
  - Next generation diagnostics and drivers
- **Energetic Materials**
  - Prediction and control of safety, initiation and performance of explosives

The implementation plan for Materials Strategy is guided by the output from the Deep Dives

- Further refine planning in the Areas of Leadership, identifying division leader champions for each area.
- Champion investments in cross-cutting capabilities in "making, measuring and modeling."
- Identify and pursue high impact program development activities
  - Enable underpinning Pu R&D in pursuit of the Nevada scaling initiative
  - Champion and seize OSTP "Functionality by Design" Opportunity
    - Critical Materials Hub (EERE)
- Pursue institutional strategies underpinning the Materials Strategy
  - Continue the strong connection with the LDRD Investment Strategy
  - Establish the "Institute for Controlled Functionality"
- Propose facility/safety basis improvements
  - Implement He recovery for condensed matter physics experiments.
  - Identify an essential activity where risk aversion limits progress (e.g. the inability to perform Pu experiments at pRad) and find a solution.

Champion investments in cross-cutting materials capabilities

Leverage MaRIE-relevant investments called out in the Deep Dives

- **Making**
- New facilities for materials chemistry, located in MSC complex
- Synthesis capability of Pu-242 for small scale experiments
- Plan for a flexible fabrication capability

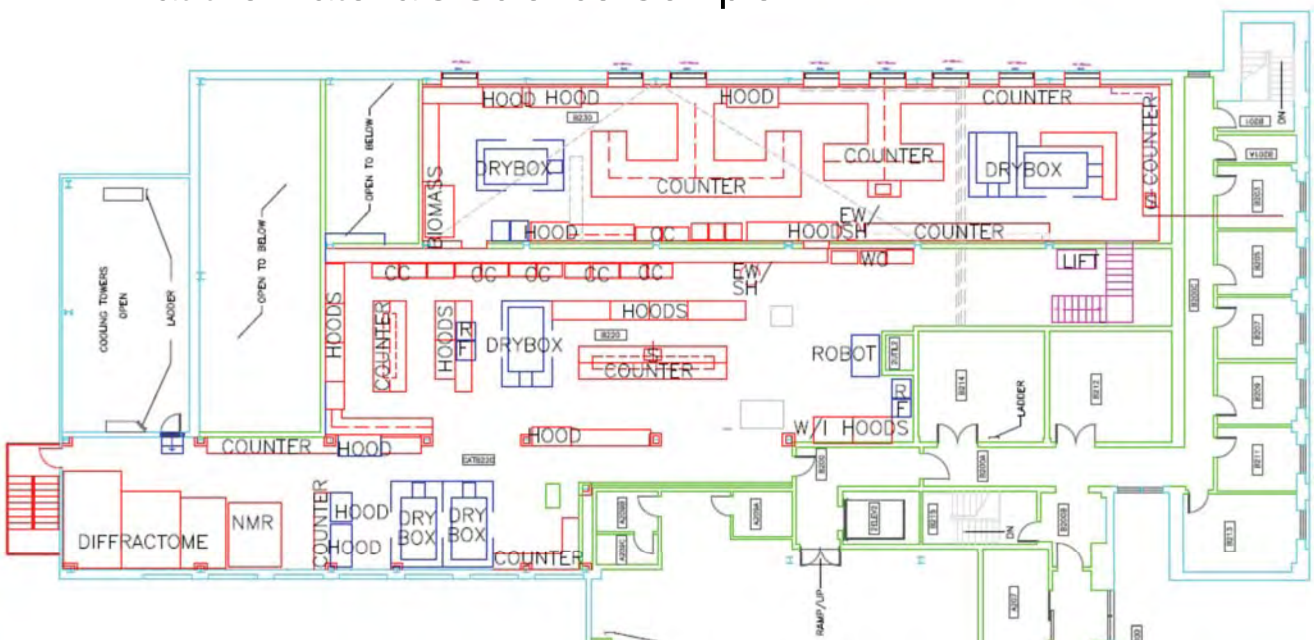
- **Measuring**
- Advance frontier microscopy capabilities
- Enhance ultrafast x-ray science at LANL and external user facilities
- Develop in situ synthesis and processing diagnostics

- **Modeling**
- Pilot projects in co-design intimately connected to experiments
  - Structural materials—underway in FY11
  - Functional materials—at a more preliminary stage

The new MSL infill lab will enable an integrated materials chemistry capability on the pathway to MaRIE and M4

Provides a molecule-to-materials capability

- In situ analysis during synthesis
- Molecular control of the surface interface critical to corrosion
- Synthesis of novel composite materials through integration with other techniques at the Materials Science Complex



~5000 sq ft

21 hoods

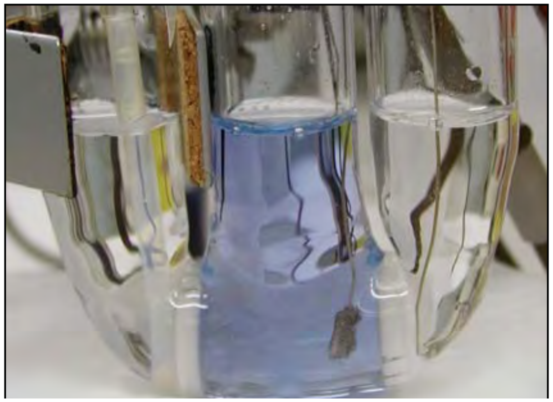
6 dry boxes

Co-located diagnostics for *in situ* characterization during synthesis and measurement of gas-solid interfacial interactions

Another objective is the establishment of a processing capability for small-scale R&D using Pu-242

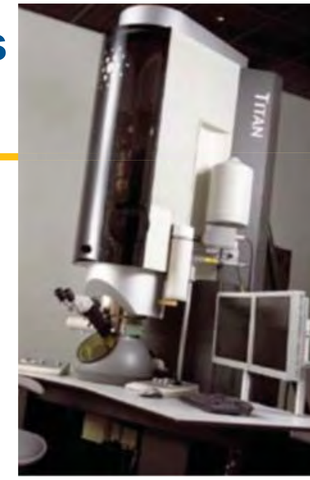
- By focusing on unique, low-specific activity isotopes like Pu-242 the whole nature of Pu research can be dramatically improved:
  - Lower security and lower hazard levels than Category I, II, or III facilities (reduced costs and bureaucracy).
  - The possibility of not having the specimens change during measurements.
  - Opportunity to engage academic collaborators and make better use of national user facilities for Pu research.
- Space in radiological facilities has been identified (TA-35, Bldg 455)
- New LDRD efforts have been initiated (reserve & LDRD-DR proposal).

Glovebox line acquired for Pu-242 effort



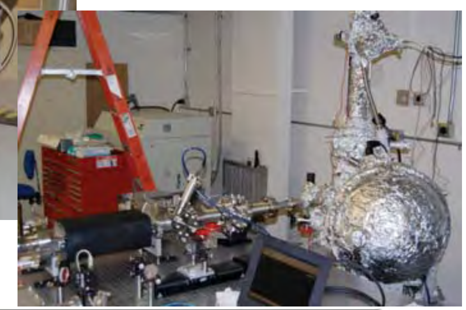
Aqueous electrochemical cell showing Pu(III) formation at Cathode

Development of advanced characterization techniques for implementation of the Materials Strategy are also essential for MaRIE



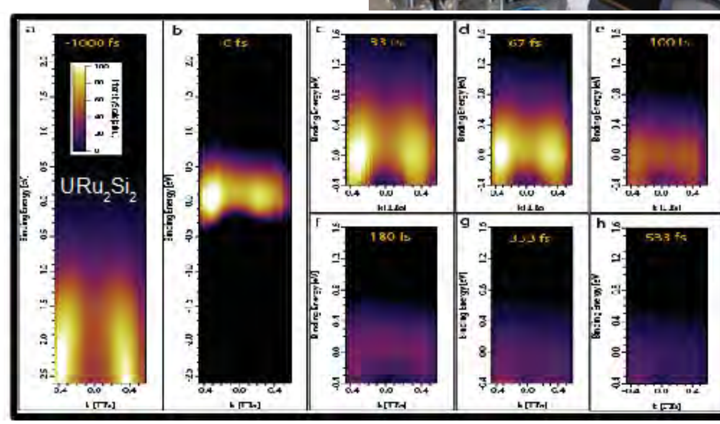
Frontiers of microscopy

- Upgraded Electron Microscopy facility
  - Progress on funding model & capitalization plan
- MaRIE-relevant LDRD projects in instrumentation & analysis



Ultrafast x-ray science

- Time-resolved ARPES based on high harmonic generation (30 fs,10-100 eV)
  - Studies reveal origin of hidden order phase in URu_2Si_2
- Development of ultrafast x-ray magnetic dichroism spectroscopy
- Benchmarking theory with experiments in coherent diffractive imaging
- Dynamic diffraction studies at APS
- Involvement in LCLS:
  - Materials in Extreme Conditions station



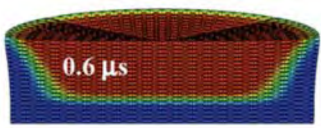
Pilot projects in codesign have been implemented, coordinated by MaRIE leadership

Control of Microstructure

Symmetry, first order transitions, tricritical behavior, hysteresis, kinetics & granularity influence the presence of retained phases under pressure in Zr, Ti. Can we control conditions that inhibit their formation ?

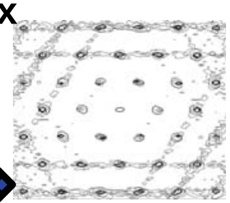
ADVANCED SIMULATION & COMPUTING

ExMatEx



IS&T
Texture analysis
--model selection
--extremal statistics
--data mining

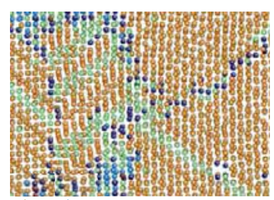
X-ray diffraction
TEM, SEM
VISAR
Neutron diffraction



Synthetic Diffraction

Phase Transformation
Twinning
Damage

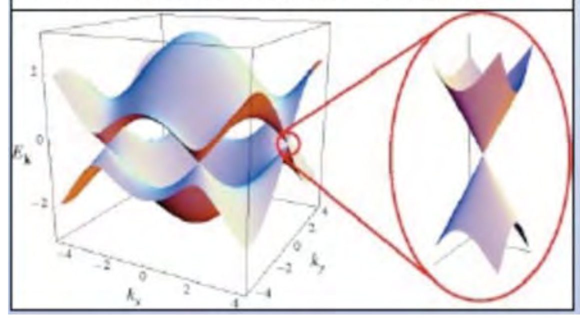
Plasticity

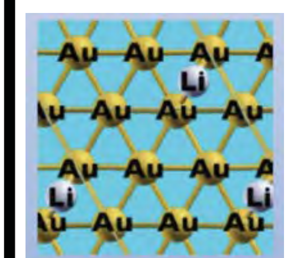


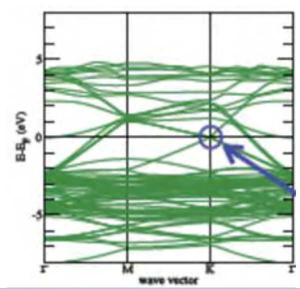
Molecular Dynamics

Designing Functional Materials

Gold clusters with up to 13 atoms have planar geometries -- can we "dope" them to create a stable 2D membrane with a Dirac cone?





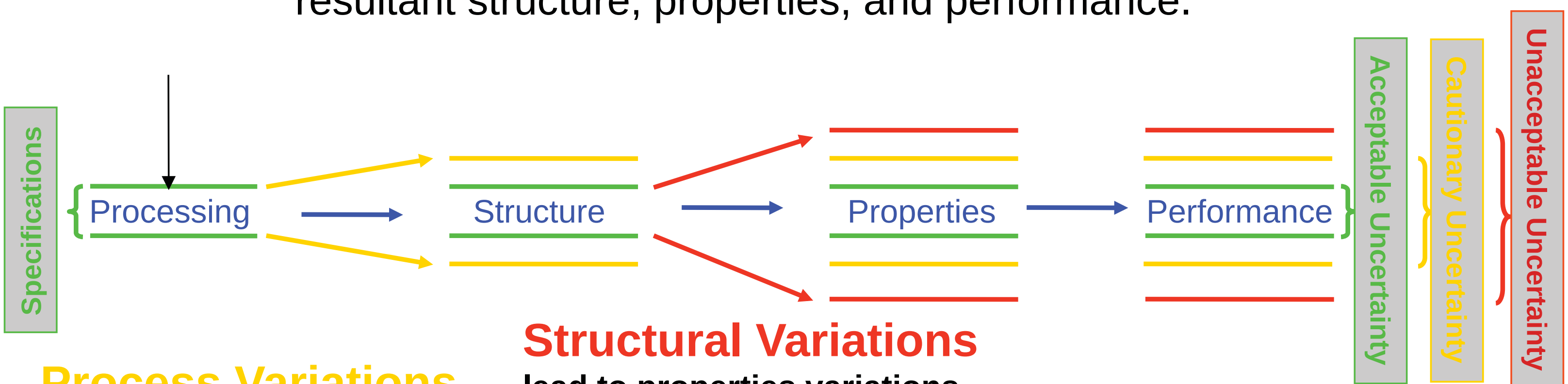


Density functional theory calculations* predict that adding a lithium atom to the membrane (one for every six gold atoms) gives the electronic structure a Dirac cone.

Program development opportunity: Enabling Pu science underpinning the Nevada scaling experiment—process aware manufacturing

Established Manufacturing Process

Known performance and margin based on well-known causal relationship between processing specifications and resultant structure, properties, and performance.



Structural Variations
lead to properties variations.

Process Variations
lead to structural variations.

Variations (intentional or not) propagate through these casual relationships and must be understood to effectively manage **impact** to **performance uncertainty**.

Establishment of the ‘Institute for Controlled Functionality’ will enhance the internal and external visibility of LANL’s materials community

Functions of the Institute:

- Coordinate activities across LANL’s materials community
  - Integration/partnership with materials-centric user facilities.
- Promote a sense of community among materials researchers and facilitate the formation of interdisciplinary teams
- Outreach to the international materials community through high visibility workshops and a vibrant visitors program.

Essential elements:

- Patterned after the (former) Center for Materials Studies (CMS) or (current) Center for Nonlinear Studies (CNLS)
- Fulltime Director with administrative support
- Dedicated space within the materials complex
- Postdoc program focused on areas of leadership
- Externally-visible workshops and visitor program

Materials Strategy and its implementation is well-connected to other LANL strategic planning efforts

- **Consistency and leverage with other Institutional Pillars**
  - Science of Signatures
  - Information Science and Technology
- **Weapons Science Strategy**
- **MaRIE**
- **Energy Strategy**
  - Materials and concepts for clean energy
  - Nuclear energy

- **Smaller scale strategic efforts align with the Materials Strategy**
  - Plutonium strategy—establishment of Pu-242 capability
  - Laser strategy
    - Resulting from Electrodynamics and Accelerators Capability Review

Next steps for the Materials Strategy Implementation Plan

- Develop a plan for ‘investment’ in people
  - Address retention and recruitment issues
  - Hold planning session focused on mentoring, career development, succession planning, awards, etc.
- Continue to present and discuss plan with stakeholders
  - Town halls for materials community
  - Materials Guiding Coalition, Division and Directorate Councils with an emphasis on their role in implementation
  - Targeted discussions with Fellows, Early Career Scientists, Postdocs
  - Discussion with programs: program development opportunities
- Incorporate feedback from stakeholders and MCR committee
- Write implementation document

MaRIE, an experimental facility concept for revolutionizing materials in extreme environments

J.L. Sarrao (SPO-SC)

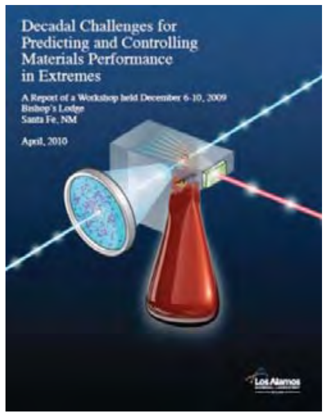
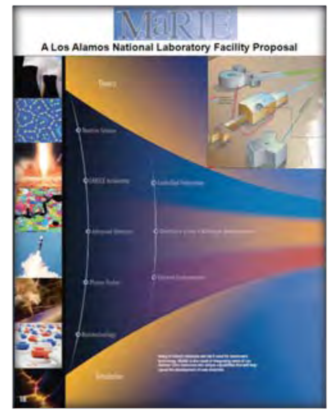
MaRIE, for Matter-Radiation Interactions in Extremes, is Los Alamos National Laboratory's facility concept for addressing decadal challenges in materials, especially in extreme environments, through a focus on predicting and controlling materials microstructure. MaRIE will be an international user facility and will enable unprecedented in situ, transient measurements of real materials in relevant extremes, especially dynamic loading and irradiation extremes. Concurrent advances in multiscale modeling and computational resources hold great promise for rapid progress toward these goals. To achieve this vision, MaRIE will construct a high-energy, low-average-intensity source of x-ray photons (pre-conceptually, a 50-100 keV XFEL) and couple it to an existing high intensity proton linear accelerator (800 MeV at 1 MW) through three measurement halls: the Multi-Probe Diagnostic Hall (MPDH), the Fission-Fusion Materials Facility (F^3), and the Making, Measuring and Modeling Materials Facility (M4). MaRIE will build upon existing investments at LANSCE, in LANL's materials infrastructure, and through national user facilities (e.g., Lujan Neutron Scattering Center, National High Magnetic Field Laboratory and Center for Integrated Nanotechnologies). MaRIE also leverages current and planned investments such as the Materials Test Station (MTS), a fast-fission irradiation capability being developed by the Office of Nuclear Energy.

MaRIE, an Experimental Facility Concept for Revolutionizing Materials in Extreme Environments

John Sarrao

UNCLASSIFIED

In response to the Dept's guidance, we are developing a pre-conceptual design proposal for MaRIE

Science Need	Facility Definition	Pre-conceptual Proposal
		
(2009)	(2010)	(2011)

Today

Current activities with NNSA and NE are key steps down the path:

LANSCE→ Linac Risk Mitigation → MTS → MaRIE

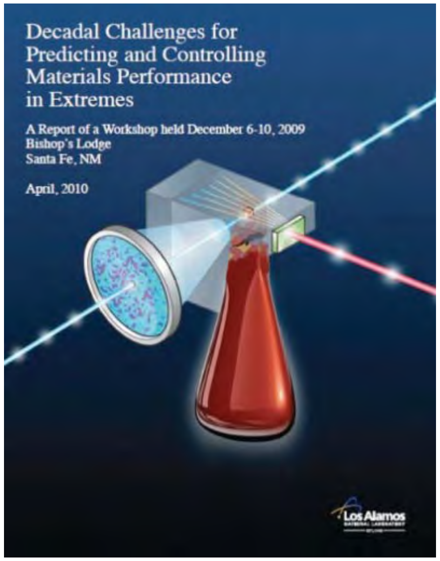
We anticipate NNSA call for large scale science facility concepts asap

Outline

- **Why MaRIE?**
- **What MaRIE?**
- **Advancing the science of MaRIE**
  - “MaRIE Program” including partners
- **Delivering a proposal for MaRIE**
  - Requirements → Scope, Schedule, Budget

Materials research is on the brink of a new era – moving from observation of performance to control of properties

- **The confluence of improved experimental capabilities (e.g. 4th generation light sources, controlled synthesis and characterization, …) and simulation advances are providing remarkable insights at length and time scales previously inaccessible**



New capabilities will be needed to realize this vision:

In situ, dynamic measurements

simultaneous scattering & imaging

of well-controlled and characterized materials

advanced synthesis and characterization

in extreme environments

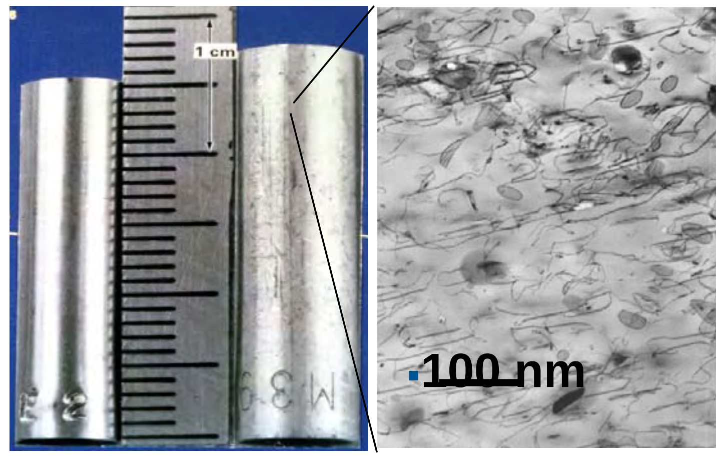
dynamic loading, irradiation

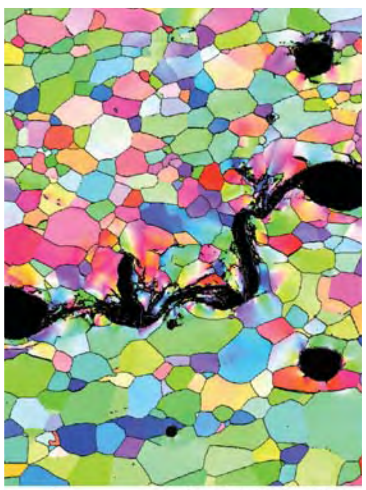
coupled with predictive modeling and simulation

materials design & discovery

The "micron frontier:" Bridging the gap between atomic understanding and bulk performance

~ 1 μm is the domain of defect consequences and microstructure interactions that drive materials strength, damage evolution, etc.

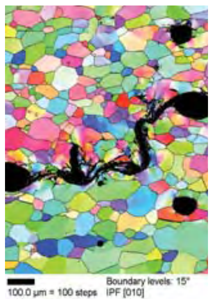




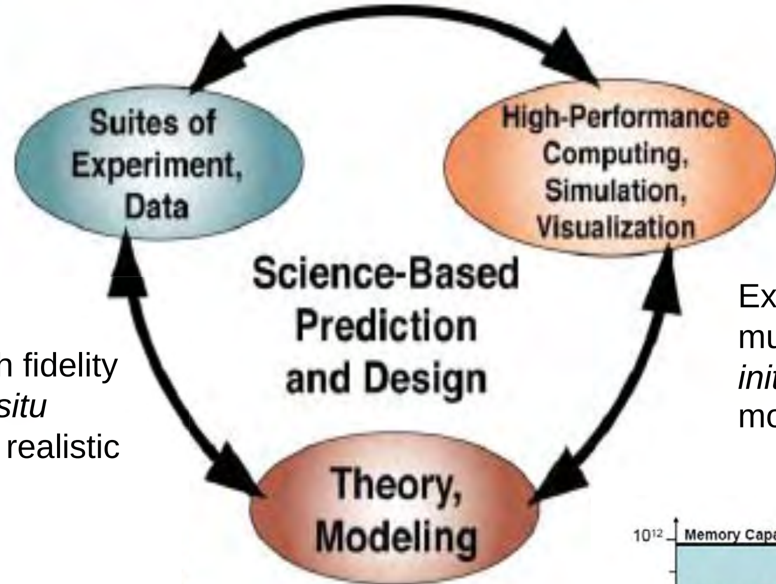
100.0 μm = 100 steps Boundary levels: 15° IPF [010]

Dynamic, stochastic processes in extreme environments dominate the phenomena that we do not understand

Next generation simulation capabilities and experimental tools will enable discovery science at the micron frontier

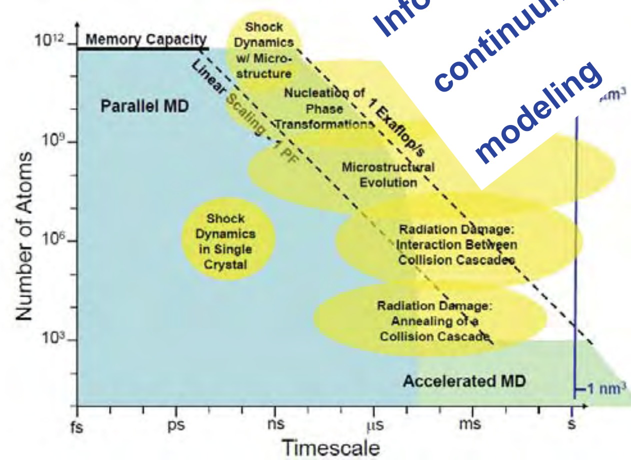


Controlled fabrication, high fidelity characterization, novel *in situ* diagnostics, generation of realistic extreme environments, …



Exascale computing, multi-scale, multi-physics simulation tools, *ab initio* methods applied to larger, more complex materia'

Multi-scale approaches to connect fundamental scales to bulk properties, defect generation and evolution, …

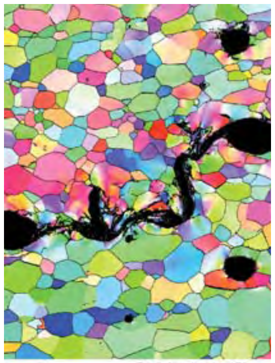


MaRIE will address problems central to Department of Energy missions in energy, science, and security

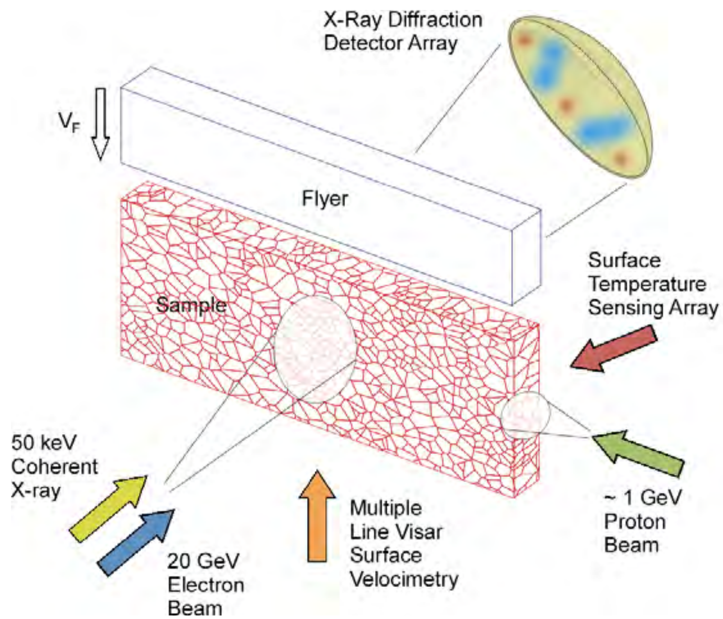
- What are the consequences of materials failure for weapons performance?
- How do we accelerate the certification of materials to enable a nuclear renaissance?
- Can we predict and prevent materials damage?
- Can we discover by design materials to perform in unprecedented irradiation extremes?
- How do we predict and control microstructure for designed materials performance?
- Can we design and synthesize new materials with controlled functionality?

Example: Predicting and preventing materials damage

Understanding the role of microstructure-based heterogeneity evolution in material damage



<u>The goal</u> :- Predict dynamic microstructure and damage evolution



+

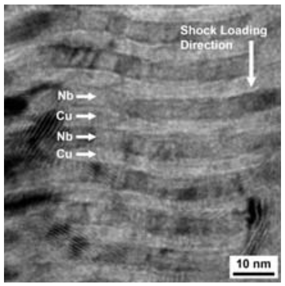
<u>The first experiment</u> :- Multiple, simultaneous dynamic in situ diagnostics with resolution at the scale of nucleation sites (< 1 µm; ps – ns)

<u>The model</u> :- Accurate sub-grain models of microstructure evolution coupled to molecular dynamics

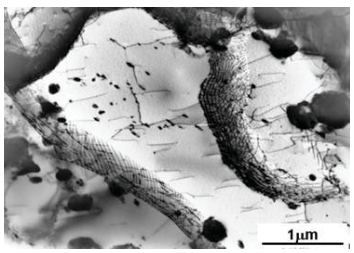
Team includes: Curt Bronkhorst et al. (LANL, UK AWE, BYU, CalTech, Ohio State, …)

Example: Prediction and control of microstructure for designed materials performance

Understanding the role of interfaces in strain evolution

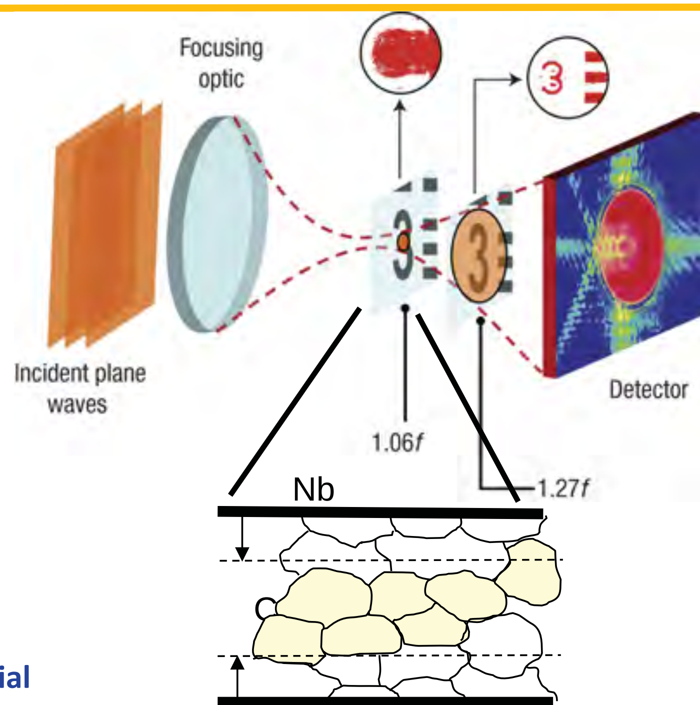


Nano laminates

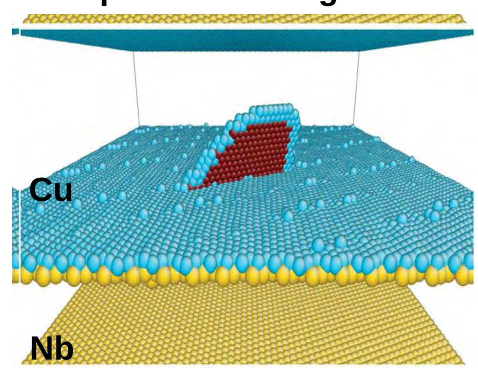


ODS steel

<u>The goal</u> : Predict interfacial microstructure for increased strength and irradiation resistance



<u>The first experiment</u> : 3-D movies of dislocation dynamics in materials at buried interfaces, micron field of view with focusing at nm resolution



<u>The model</u> :Advanced M²S with micron scale, multigranular predictions

Team includes: Nate Mara et al. (LANL, ANL, CMU…)

MaRIE builds on the LANSCE facility to provide unique experimental tools to meet this need

First x-ray scattering capability at high energy and high repetition frequency with simultaneous charged particle dynamic imaging

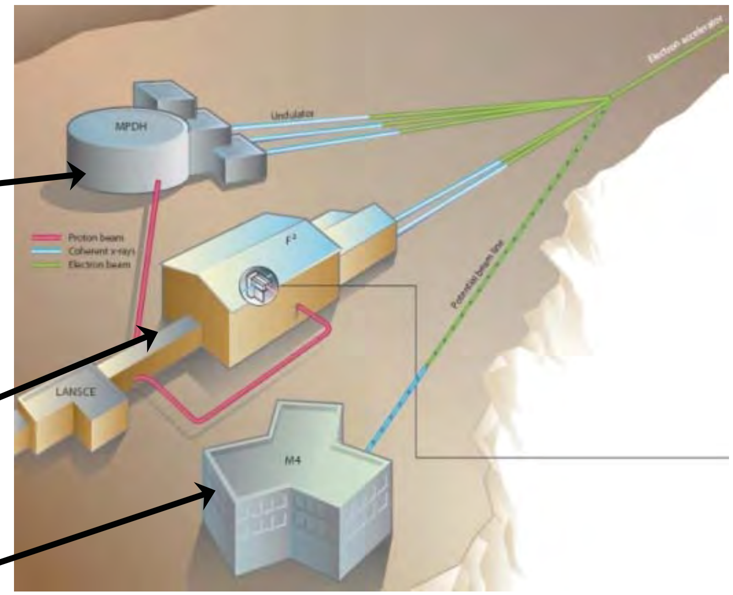
(**MPDH**: Multi-Probe Diagnostic Hall)

Unique in situ diagnostics and irradiation environments beyond best planned facilities

(**F³**: Fission and Fusion Materials Facility)

Comprehensive, integrated resource for materials synthesis and control, with national security infrastructure

(**M4**: Making, Measuring & Modeling Materials Facility)



Unique very hard x-ray XFEL
Unique simultaneous photon-proton imaging measurements
Unique spallation neutron-based irradiation capability
Unique in-situ, transient radiation damage measurements
Unique materials design and discovery capability

MaRIE will provide unprecedented international user resources

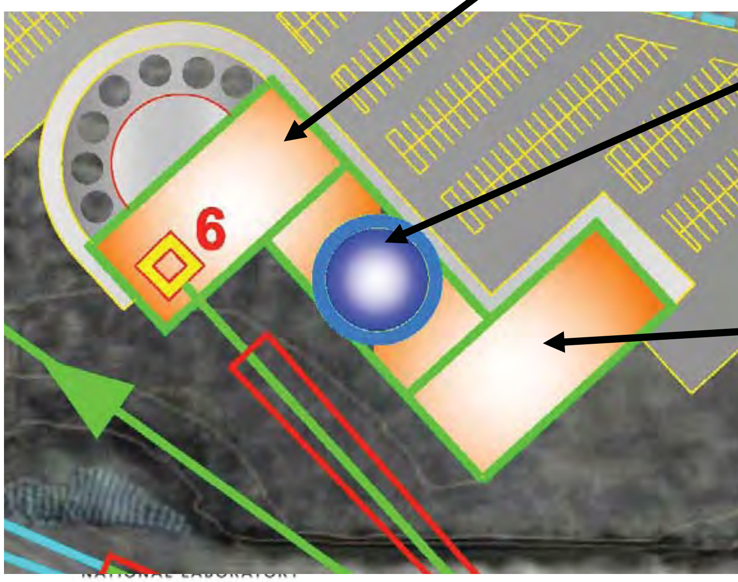
MaRIE photon needs can be met by an XFEL that is technically feasible and affordable

	MPDH	FFF		M4	
Energy/Range (keV)	50	<10 - >50	10-400	0.1-1.5	10-50
Photons per image	10^{11}	10^{11}	10^{9}	10^{9}	10^{11}
Time scale for single image	50 fs	>1 s	0.001 s	10-500 fs	50 fs
Energy Bandwidth (ΔE/E)	10^{-4}	10^{-4}	10^{-3}	10^{-4}	10^{-4}
Beam divergence	1 μrad	1 μrad	< 10 μrad	< 10 μrad	1 μrad
Trans. coherence (TC) or spatial res.	TC	TC	1-100 μm	TC	TC
Single pulse # of images/duration	100/1.5 μs	-	-	-	-
Multiple pulse rep. rate/duration	120 Hz/day	0.01 Hz/mo.	60 Hz/secs	1 KHz/day	10 Hz/days
Longitudinal coherence	yes	yes	no	yes	yes
Polarization	linear	linear	no	Linear/circular	linear
Tunability in energy (ΔE/E/time)	2%/pulse	fixed	fixed	10%/s	10x/day

- Photon energy - set by gr/cm^2 of sample and atomic number
- Photon number for an image - typically set by signal to noise in detector and size of detector
- Time scale for an image - fundamentally breaks down to transient phenomena, less than ps, and semi-steady state phenomena, seconds to months
- Bandwidth - set by resolution requirements in diffraction and/or imaging
- Beam divergence - set by photon number loss due to stand-off of source/detector or resolution loss in diffraction
- Source transverse size/transverse coherence - the source spot size will set the transverse spatial resolution, if transversely coherent then this limitation is not applicable so transverse coherence can be traded off with source spot size and photon number
- Number of images/rep rate/duration – images needed for single shot experiments/image rep rate/ duration of experiment on sample
- Repetition rate - how often full images are required
- Longitudinal coherence – 3D imaging
- Polarization - required for some measurements
- Tunability – time required to change the photon energy a fixed percentage

Through M4 Facility, MaRIE provides the directed synthesis of materials essential for defect/interface control and materials discovery

	M4	
Energy/Range (keV)	0.1-1.5	10-50
Photons per image	10^9	10^{11}
Time scale for single image	10-500 fs	50 fs
Energy Bandwidth (ΔE/E)	10^{-4}	10^{-4}
Beam divergence	< 10 μrad	1 μrad
Trans. coherence (TC) or spatial res.	TC	TC
Single pulse # of images/duration	-	-
Multiple pulse rep. rate/duration	1 KHz/day	10 Hz/days
Longitudinal coherence	yes	yes
Polarization	Linear/circular	linear
Tunability in energy (ΔE/E/time)	10%/s	10x/day

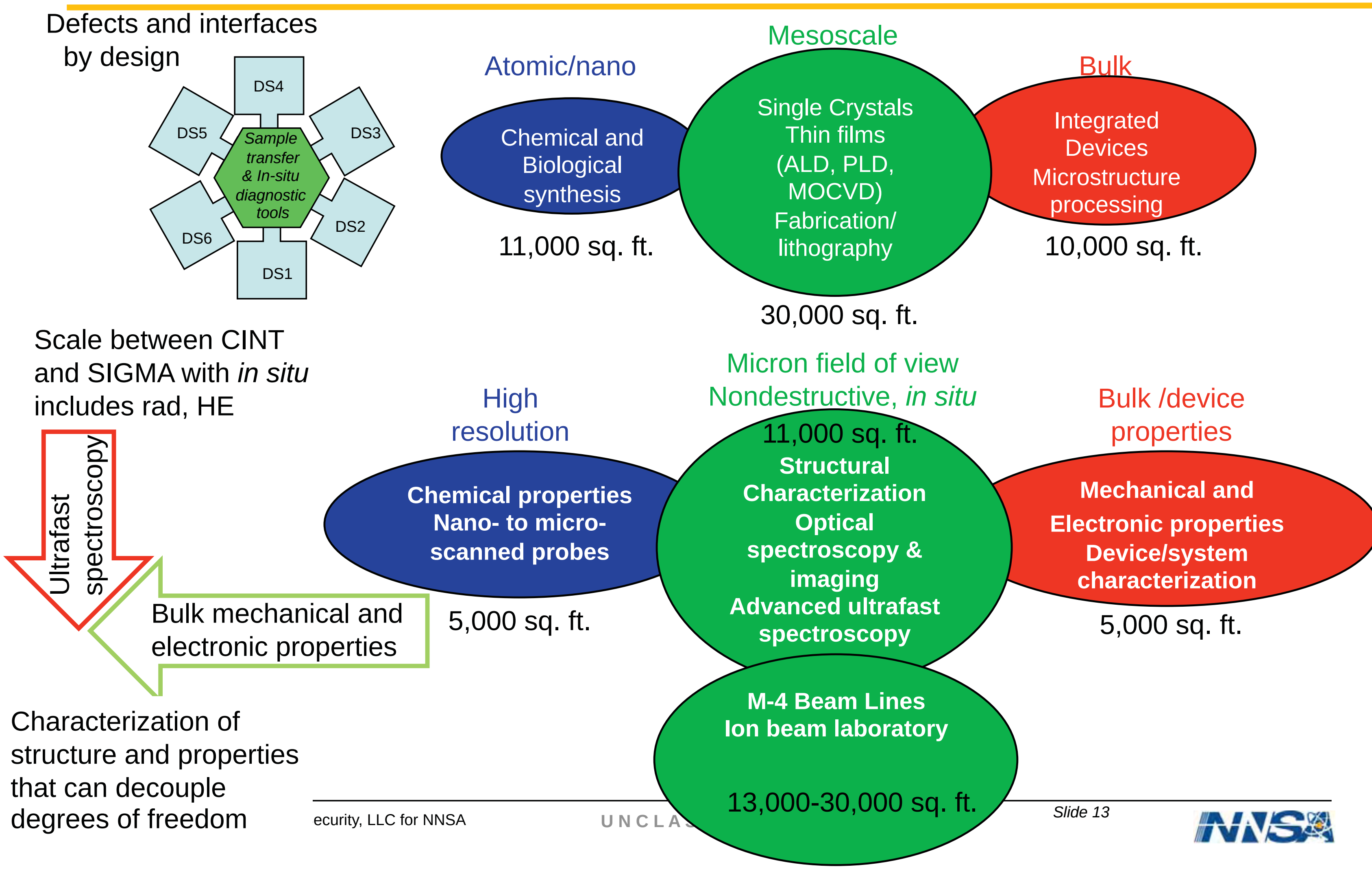


M4 XFEL end station (hard & soft x-rays)
Other Extremes (E,H, pH)
In situ synthesis probes

User Gateway
Co-design Center
Visualization Capability

Multi-scale Synthesis & Crystal Growth
Characterization
National Security Infrastructure

M4 – Integrated mesoscale synthesis and nondestructive characterization with micron field of view



FY11 LDRD Reserve investments targeted on materials in extremes are accelerating science underlying MaRIE first experiments

XFEL physics

Coherence Effects in x-ray Diffraction Imaging — Quinn Marksteiner/John Barber

Control of XFEL-Radiation Focusing through Electron-Beam Manipulation — Kip Bishofberger

APS expts

Synchrotron X-ray Laue Diffraction and Phase Contrast Imaging of Fe and Explosive Simulants under Shock Loading — Shengnian Luo

In-Situ Probing Monitoring of Microstructure Evolution During Annealing of Radiation Damage with High Energy Synchrotron Xray Diffraction — Donald Brown

Three Dimensional Quantification of Metallic Microstructures in the Presence of Damage — Curt Bronkhorst

Advanced pRad

Achieving the Ultimate Spatial and Density Resolution of 800 MeV Proton Radiography — Alexander Saunders

Synthesis science

Developing and synthesizing epitaxial nanocomposites with controlled defect landscapes and desired functionalities — Quanxi Jia

Fluid Flow Imaging of Alloy Melts and In-situ Fundamental Solidification Experiments at Temperature Extremes — Amy Clarke

Microstructure Analysis for Extreme Events: A Stochastic Modeling Framework for Microstructure Datasets — John Bingert

Accelerating Materials Certification — John Sarrao

“Accelerating Materials Certification” Project

We are pursuing

i) the **co-design of micro-mechanical models** to improve the predictive capability of engineering analyses into the regimes of high-pressures and strain rates,

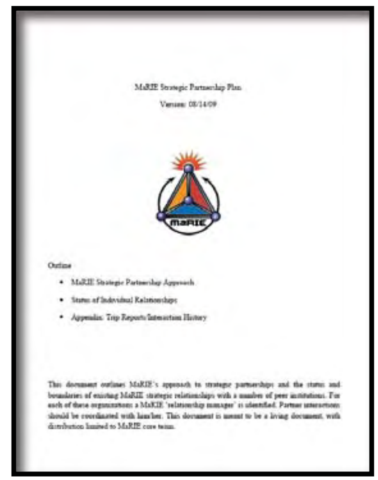
ii) utilization of **ion beam irradiation** as a means of **accelerating materials damage** with the goal of realizing improved certification models of relevance to nuclear energy,

iii) ultrafast in-situ, transient measurement approaches to separate coupled degrees of freedom essential to predicting materials functionality, and

iv) synthesis strategies for tailoring materials properties based on **near-real-time diagnostic feedback**.

Consistent with our roadmap, we are fostering strategic partnerships

- National laboratories
  - -LLNL & SNL: nuclear weapons
  - -ORNL & INL: nuclear energy
  - -ANL & LBNL & SLAC, and FNAL: photon and proton accelerator development
  - -SRNL, PPPL, BNL, PNNL, JLAB: ongoing outreach
  - -Ames, NREL, NETL: initial interactions
- Universities
  - -Key partners in community outreach/first experiments teams
    - -typically not stewards of large-scale facilities
- Industry
  - -Focus on consortia advocacy
    - -pillar specific; lead with F^3



MaRIE Strategic Partnership Plan

Version: 08/14/09

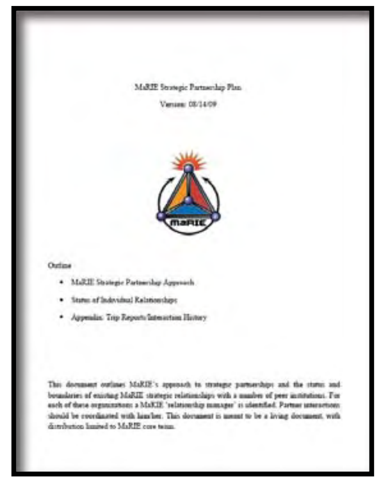
Outline

- MaRIE Strategic Partnership Approach
- Status of Individual Relationships
- Appendix: Trip Reports/Interaction History

This document outlines MaRIE's approach to strategic partnerships and the status and boundaries of existing MaRIE strategic relationships with a number of peer institutions. For each of these organizations a MaRIE 'relationship manager' is identified. Partner interactions should be coordinated with him/her. This document is meant to be a living document, with distribution limited to MaRIE core team.

A short history of recent partnership activities: Focus on national labs

- National Laboratories: Recent activities
  - -LLNL & SNL: “MaRIE Days” at LLNL & SNL with documented follow-up plans
  - -SLAC: XFEL and Accelerator r&d partnering
  - -ANL: DC-CAT and other APS-U end stations, “Materials for Energy”
  - -BNL: Reciprocal visits planned – May and June
- Universities
  - -Key partners in community outreach/first experiments teams
    - -typically not stewards of large-scale facilities
- Industry
  - -Focus on consortia advocacy
    - -pillar specific; lead with F^3



MaRIE Strategic Partnership Plan

Version: 08/14/09

MaRIE

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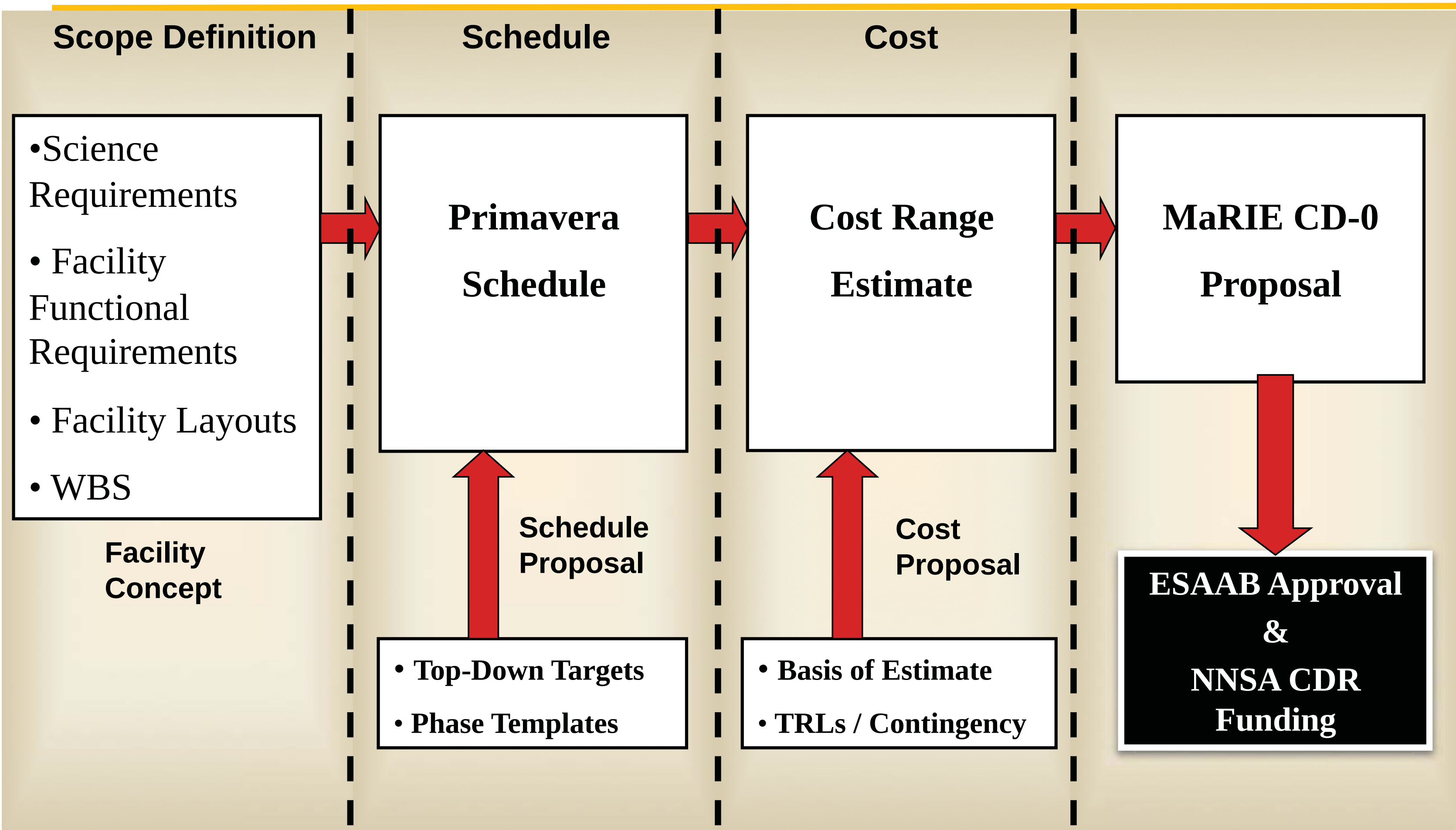
Applying appropriate amount of PM rigor to CD-0 Plan



Summary

Science-driven mission need leads to integrated facility needs fulfilled by MaRIE

User Driven Science → Materiel Needs → **Functional Requirements** → Alternatives Analyses → **Performance Gaps** → Preferred Alternative & Roadmap → **Facility Concept**

User Driven Science

Dynamic Extremes
- Microstructure Evolution
- Stochastic Explosive Microstructure & Detonation
- Fluid/Mineral Interactions in
- 3-D Measurements of Turbulent

Radiation Extremes
- Irradiation Stability of Structural Nanocomposites
- Fission Gas Bubble & Swelling in UO_2 Nuclear Fuel
- Mechanical Testing of Structural Materials in Fusion/Fission Environ.
- Measurements of Temperature, Microstructure & Thermal Transport
- Rad Damage in Passive Oxide Films & its Influence on Corrosion

Control of Complex Materials & Processes
- Understanding Emergent Phenomena in Complex Materials
- Developing Practical Superconductors by Design

Energy Conversion & Storage
- Achieving Practical High-Density Energy Storage Through New Support/Catalyst Electrode Systems
- Solar Energy Conversion w/ Functionally Integrated Nanostructures

Process-Aware Materials Performance
- Nanostructured Ferritic Alloys
- Exploring Separate Effects in Pu

Functional Requirements

Environments
- Dynamic pressure <200 GPa
- Strain rate = 10^1–10^7 s^{-1}
- Temperature = 77–2000 K
- High Explosives < 30 g
- Pu isotope samples < 3 mm thick
- Irradiation rate < 35 dpa/fpy
- He(appm)/dpa ratios: 0.1-1, 9-13
- Irrad Volume: 0.5 l @ >14 dpa/yr

Measurements
- Scattering
  - Defects: 1 nm res over 10 um
  - Stress: 1-2 um res over 100 mm
  - Lattice Strain: 10 nm res in 3D
- Density Imaging
  - 0.1-1 nm, <1-ps res over 10 μm
  - 10 nm, <1-ps over 50 μm
  - 0.1-1 um, < 0.3 ns over 0.1-1 mm
- Spectroscopic
  - 3D chemistry mapping w/ 1μm res
- Themo-Physical Measurements
  - Temperature: 1 μm res
  - Thermal Conductivity w/ 1 mW/m-K res

Synthesis with Characterization
- Organic, inorganic, biomaterials incl nanomaterials, HE & actinides
- Thin films with buried interface characterization

Performance Gaps

50 keV coherent x-ray source with 10^{11} photons per macropulse focused to 1-200 μm

Dynamic charged particle imaging with 20-GeV electrons

Tunable ultrashort x-ray source for excitation: 5-35 keV, 100 fs, focused to 10 nm

Ultra short pulse lasers for spectroscopy: THz (2 meV) to VUV (6 eV)

MW fast neutron source with $2x10^{15}$ n/cm2-s and >4000 h/yr operation with < 10 beam trips per day over 1 min

Crystal growth with control of impurities & defects during and after fab

Deposition Lab w/CVD, PVD, evaporation, ion beams

Nanofabricaiton Lab w/ lithography, dry & wet etch, thermal processing

Characterization Lab w/ SEM, FE-SEM, AFM, SALVE, ion beams

Data Visualization Lab w/ 1MB-10TB available per expt.

Facility Concept

Build upon $B LANSCE site credits by adding:

- **Accelerator Systems**
  - ➢ Electron Linac w/XFEL
  - ➢ LANSCE power upgrade
- **Experimental Facilities**
  - ➢ Multiprobe Diagnostic Hall (MPDH)
  - ➢ Fission-Fusion Materials Facility (F-cubed)
  - ➢ Making, Measuring, & Modeling Material Facility (M4)
- **Conventional Facilities**

MaRIE Reviews since May, 2010: Science "Deep Dives" & translation to project discipline

External Advisory Board May 25-26, 2010 "Shoot for CD-0 package in July 2010"

Committee Members: John Hemminger, Irvine, Chair; Charlie Baker; Mike Cappiello; George Crabtree, ANL; Roger Falcone, LBNL; Paul Fleury, Yale; Bill Frazer, UC; Bill Goldstein, LLNL; Rob Goldston, PPPL; Rus Hemley, Carnegie; Bill Herrmannsfeldt, SLAC; Arthur Kerman, MIT; Christian Mailhiot, LLNL; Thom Mason, ORNL; Cherry Murray, Harvard; Roy Schwitters, Texas; Robert Selden; Steve Zinkle, ORNL

- **F^3 Deep Dive September 13-14, 2010**
- **MPDH Deep Dive September 16-17, 2010**
- **M4 Deep Dive November 3-4, 2010**
- ***XFEL Pre-Conceptual Design Review September 12, 2010***
- ***XFEL Cost Review November 3-4, 2010***

External Advisory Board November 16-17, 2010

"We encourage the project to produce a draft strawman CD-0 package as planned. Moving this process forward efficiently will help to identify specific challenges within DOE that need to be addressed as well as opportunities that may come up."

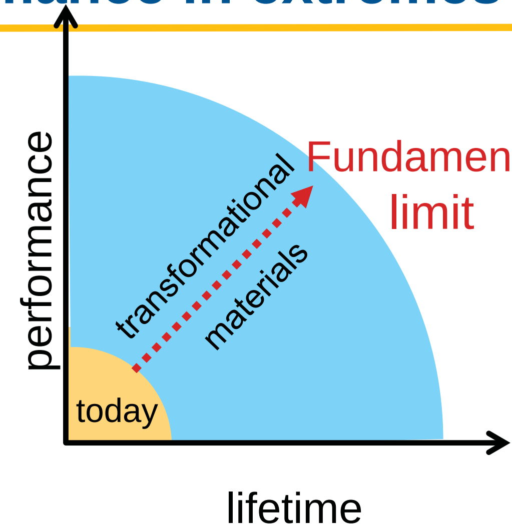
Proposal/Project Review February 22-23, 2011

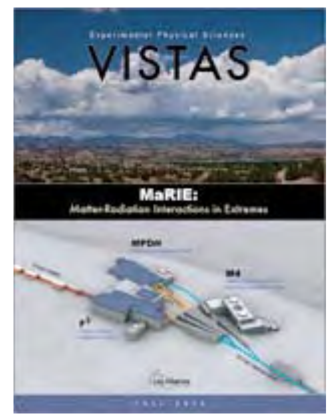
"A Los Alamos team has successfully brought the MaRIE case to a strong position for supporting DOE CD-0 requirements."

"The committee supports the LANL plan to proceed to prepare CD-0 package…package must be developed to meet needs of the NNSA client given that they will be the majority stakeholder"

MaRIE will be the first capability with unique co-located tools necessary to realize transformational advances in materials performance in extremes

- **“The micron frontier” is key to solving transformational materials grand challenges**
- **MaRIE will provide unique capabilities**
  - Accessing materials irradiation/damage extremes
  - Simultaneous in situ imaging & scattering measurements
  - Accelerating materials discovery and solutions through control of defects and interfaces
- **MaRIE provides unprecedented international user resources for the transition from observation to control**
- **Facility definition being driven by community-validated performance gaps & functional requirements**



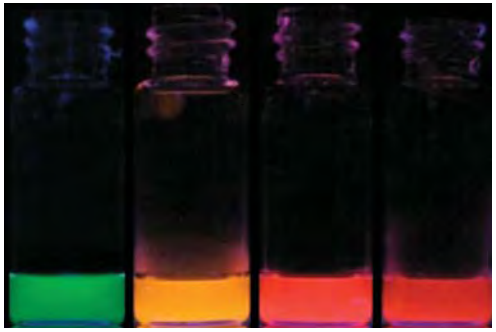


Predictive design of noble metal nanoclusters

J. Martinez (MPA-CINT)

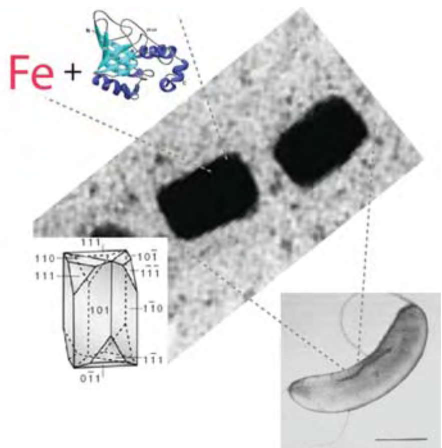
We will mimic and exploit biology to develop noble metal nanoclusters with exquisite precision (size and structure), and we will use state-of-the-art experimental and theoretical tools to develop an understanding of the electronic structure and remarkable photophysical properties of these materials. Fluorescent few-atom metal nanoclusters are collections of small numbers of gold or silver atoms with strong fluorescence emission. Nanoclusters maybe ideal for use in mission-relevant applications including the imaging of complex biological systems, the detection of biothreat agents, and the development of nanoscale materials for the harvesting and manipulation of solar energy, if efficient methods can be developed to generate clusters with ease for bioconjugation. Toward these goals, we will explore bio-inspired and traditional methods of cluster synthesis; characterize cluster morphology and photophysical properties; correlate these experimental measurements with electronic structure calculations and use these to predict ideal cluster morphology; and demonstrate the utility of the clusters for applications in biological sensing and imaging. Ultimately, through iteration between synthesis, characterization, theory, and application, we will produce a full color palette of stable, biologically compatible, luminescent nanoclusters.

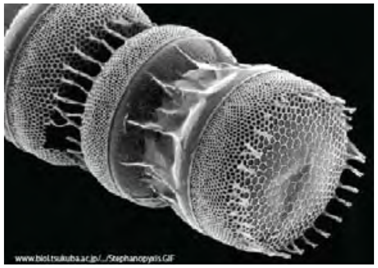
Predictive Design of Noble Metal Nanoclusters

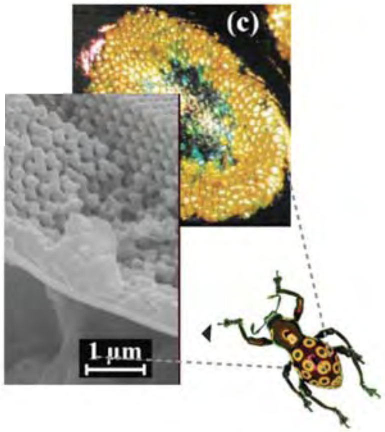


Jen Martinez (jenm@lanl.gov); Jim Werner; Sergei Ivanov; Andy Shreve; Andrei Piryatinski; Dung Vu; Sergei Tretiak; Ryszard Michalczyk; Tim Yeh; Jaswinder Sharma; Indika Arachchige; Mike Neidig; Kirill Velizhanin; Satyender Goel

Nature: a mixture of hard and soft materials





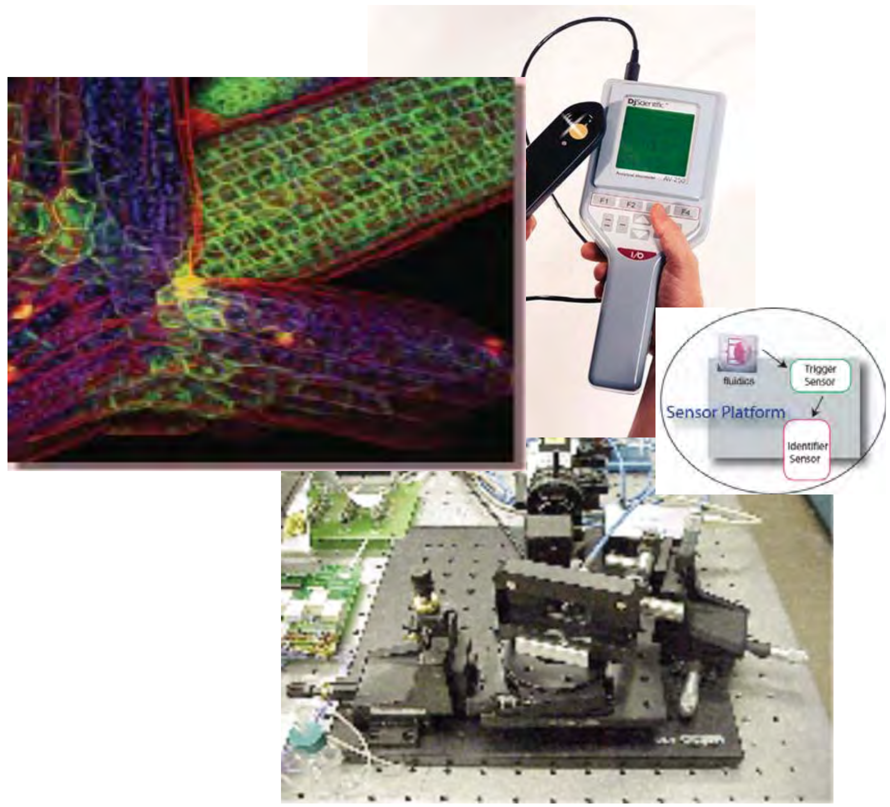


Materials Fabrication in Nature:

- **programmed** **DNA ⟶ MATERIALS**
- **precise**
  + atom-by-atom assembly from soluble inorganic precursors
- efficient and reproducible
- **materials with defined structure and properties**

Stucky/ Morse *P.N.A.S.* (1998) 95
Blakemore *Science* (1975) 190
Bartle *Phys Rev. E* (2008) 77

Sensing and imaging for disease and biothreat reduction:

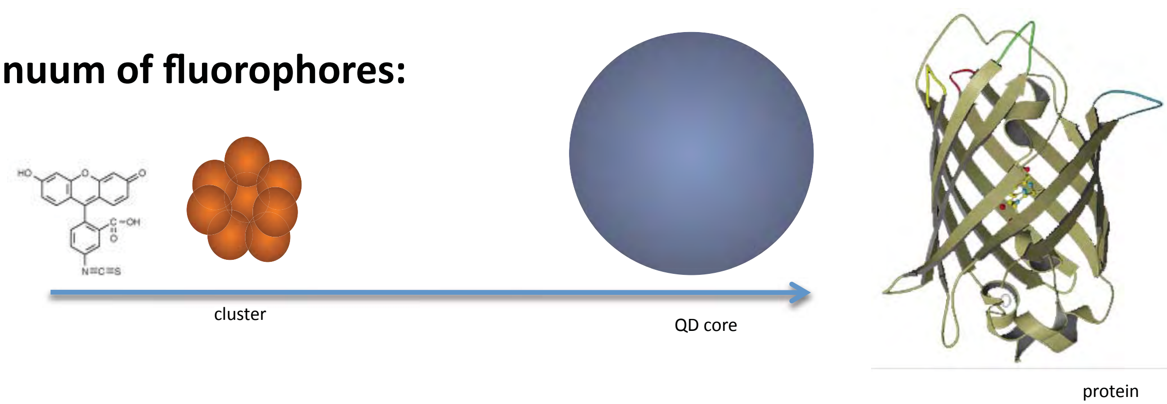


Ideal fluorophores for bioimaging:

Ideal fluorophores:

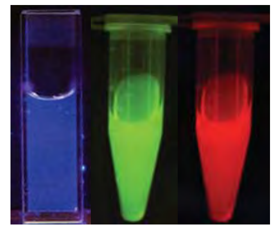
- non-toxic to cells
- smaller than biomolecules
- no photobleaching nor blinking
- short fluorescent lifetimes and high quantum yields
- easy attachment to biomolecules
- tunable (wavelength)

Continuum of fluorophores:

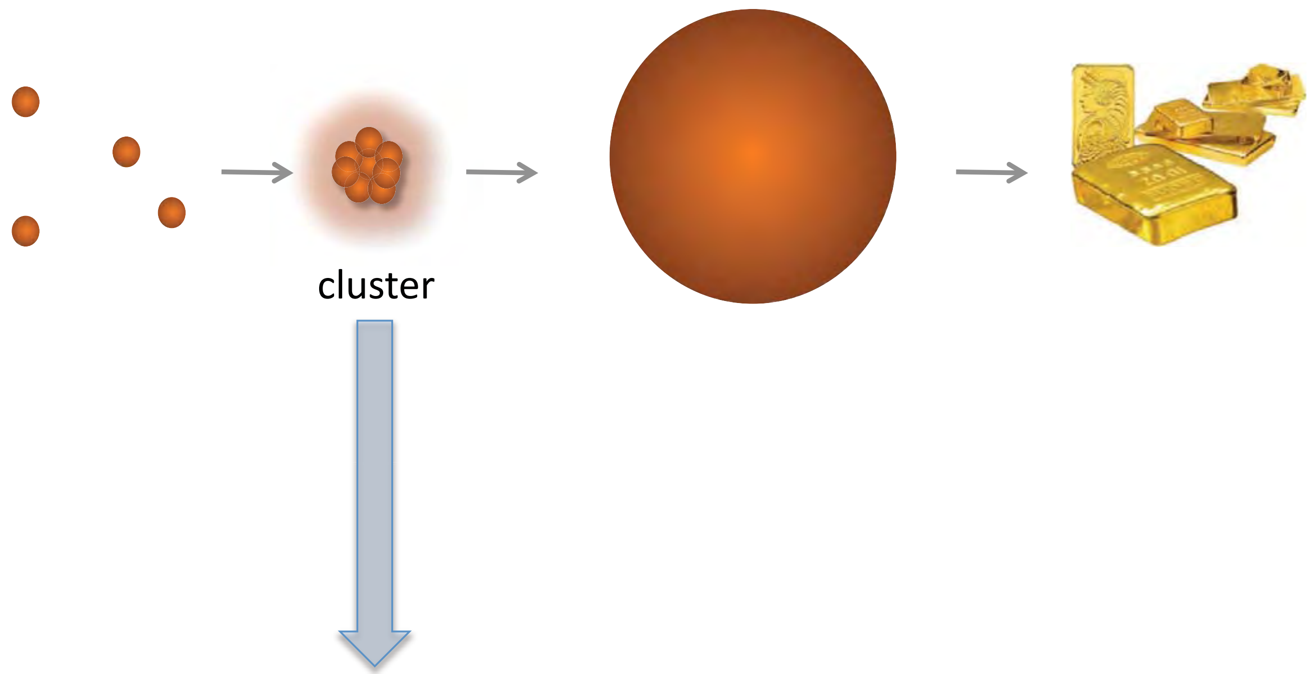


Fluorescent noble metal nanoclusters are small and bright:

-  (2-31 atoms) of gold or silver, ≤ 1.5 nm
- smaller than proteins and DNA
- fluorescent
- behave as molecular systems: discrete states and (dogma) size-dependent emission (Au)
- biologically compatible

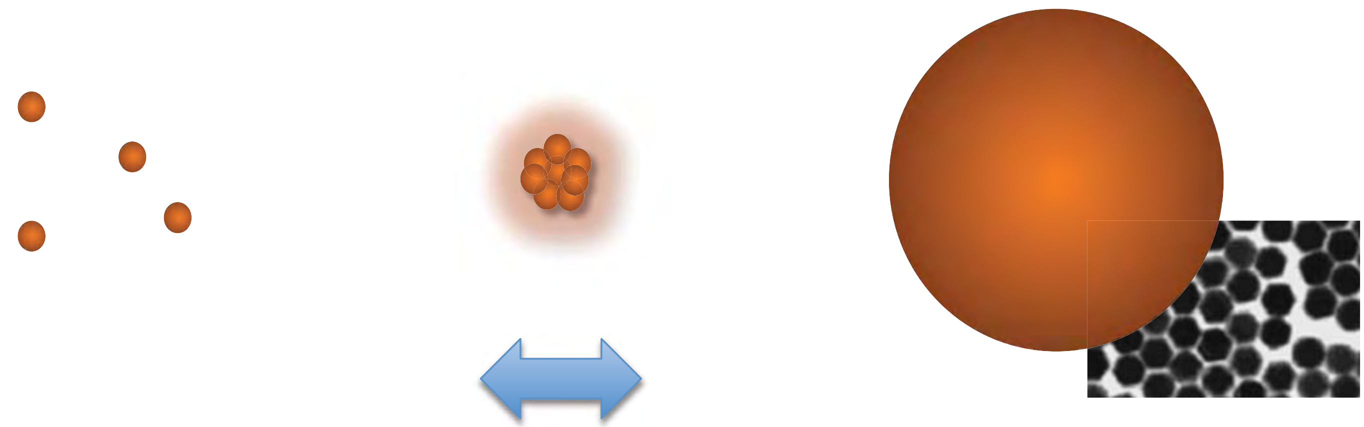


Clusters are hard to make and study:



- hard to make
- hard to physically characterize
- hard to predict (model)

Clusters are hard to make and study:



Crystal structures, NMR

TEM

Ziyou Li *J.A.C.S* (2010) 9
Nuzzo *J. Phys. Chem* (2006) 110

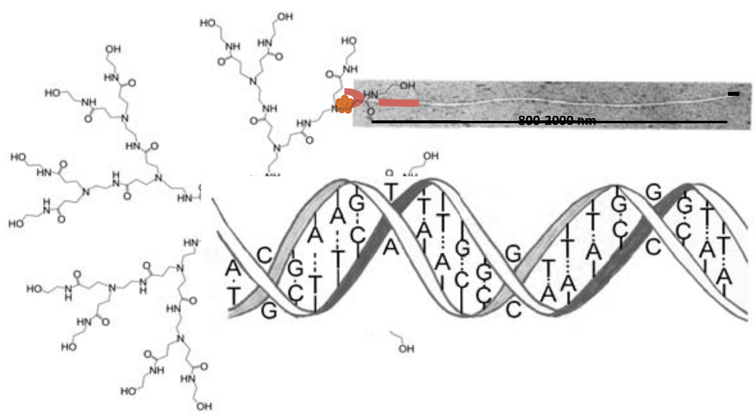
Templates stabilize and functionalize clusters:

Templates (ligands):

- provide affinity for- and organization of- gold
- prevent aggregation toward nanoparticles
- *determine the fluorescence*
- provide functionalization

Templates for <u>fluorescent</u> clusters:

- Ar matrix
- Dendrimer
- Polymers
- DNA
- Protein
- Peptides
- Small molecules



Schmid: Angew. Chem. (1993) 250
Cariati: Chem Comm (1965) 212
Murray: Acct. Chem. Res. (2000) 27
Nuzzo: J.A.C.S (2005) 812
Whetten: J. Phys Chem C (2009) 113

Fundamental grand challenges remain for ultimate application:

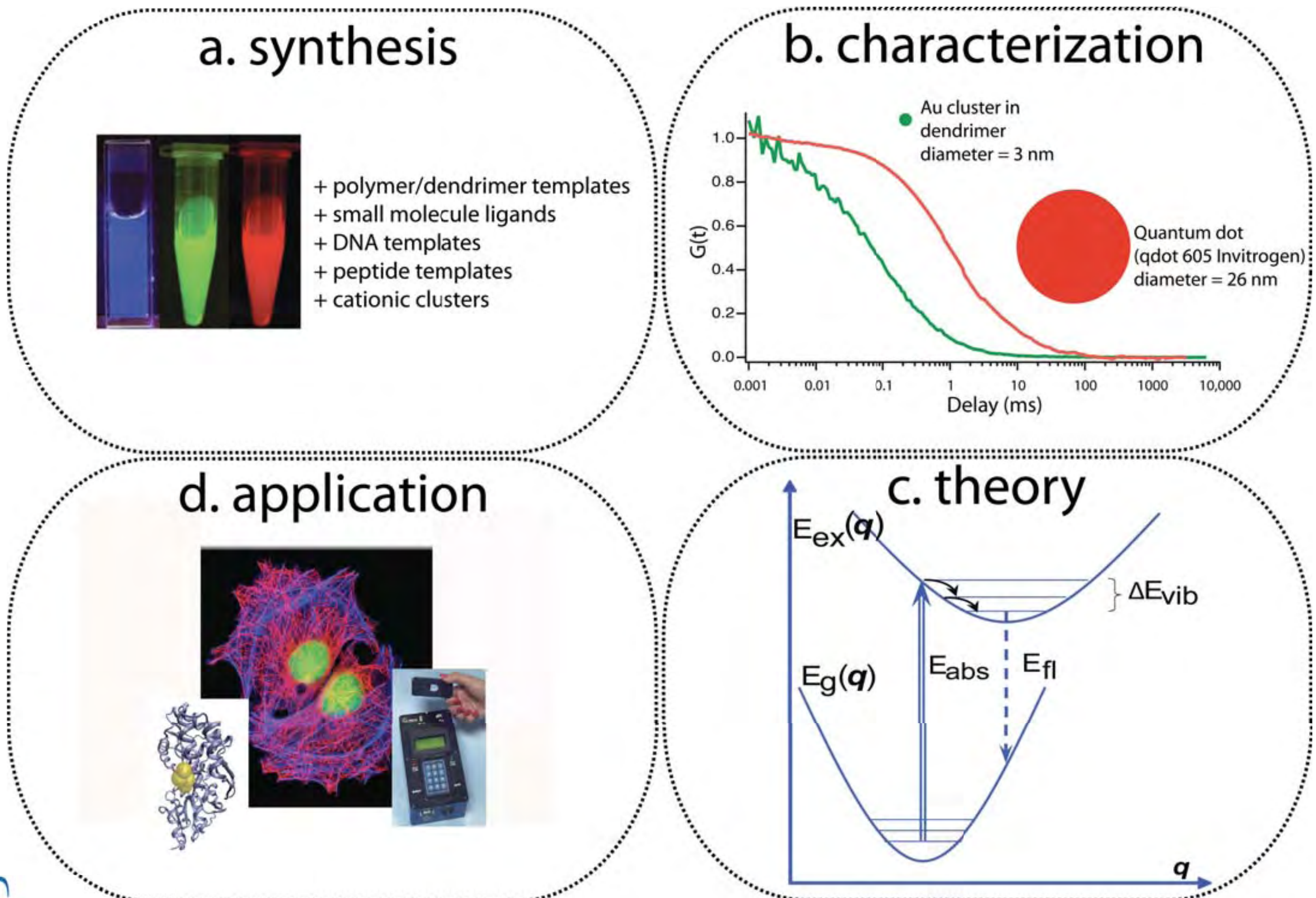
Fundamental questions:

- How to precisely control fluorescence?
  - role of ligands
  - role of charge
  - how to tune quantum yields/photostability
  - correlation of size to fluorescence
- Why are collections of atoms fluorescent? Mechanisms?
- Predictive design of nanoclusters with defined fluorescence

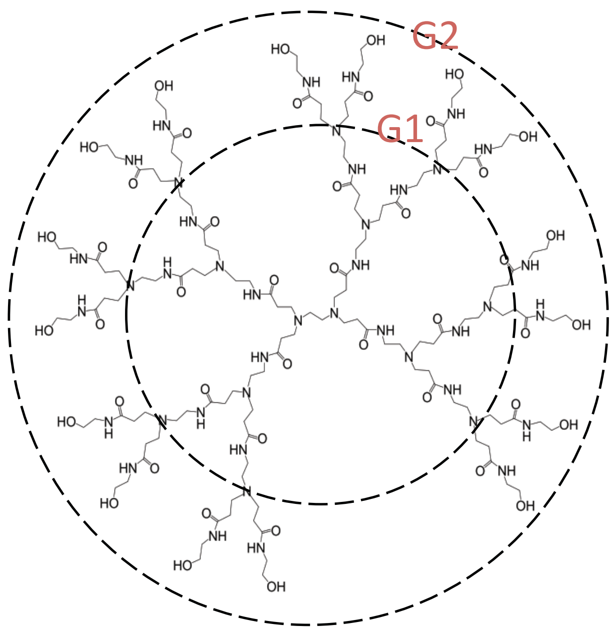
Practical "issues":

- Reproducible synthesis (new strategies/new templates)
- Synthesize palette of different colored fluorescent clusters
- Detailed photophysical characterization of clusters
- Detailed chemical/structural characterization of clusters
- Conjugation of clusters to biomolecules
- Use of clusters in sensing and imaging

Developing a full color palette of fluorescent nanoclusters:

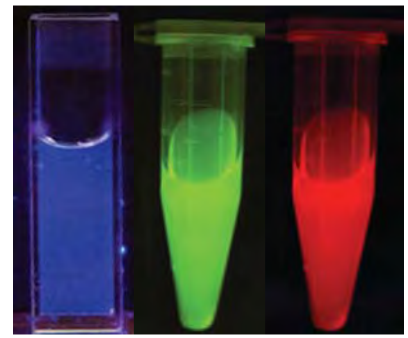


Polyamine dendrimers: gold cluster templates



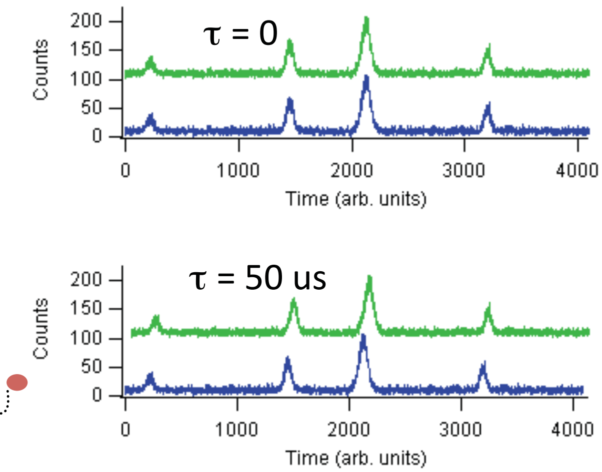
PAMAM
Dendrimer

$HAuCl_4$
$\xrightarrow{\hspace{3em}}$
37 C
+/- ascorbate



Bao, Y., et. al. "Nanoparticle free synthesis of fluorescent gold nanoclusters at physiological temperature." *J. Phys. Chem*. C 111(33), 12194-12198 (2007).
Zhong, C.,et. al. "Fabrication of fluorescent cellular probes: hybrid dendrimer/gold nanoclusters." *Mater. Res. Soc. Symp. Proc.*1007 (2007, 1007-S15-07)

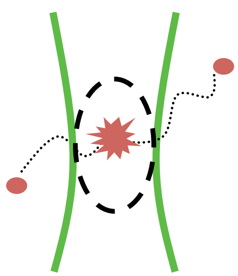
Fluorescence correlation spectroscopy: size, concentration, brightness



Amplitude: Gives Number of particles in probe volume

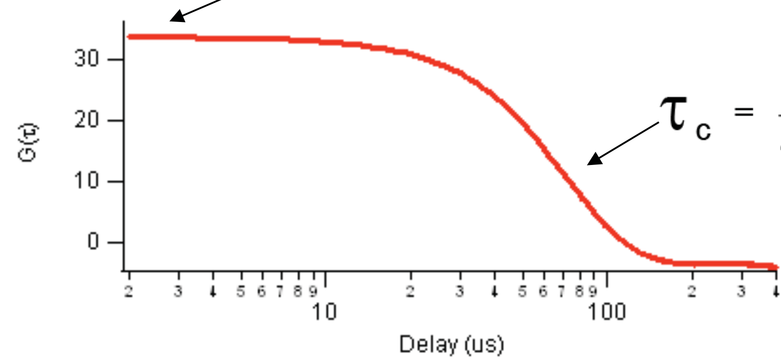
in turn, gives a measure of
- synthetic yield
- extinction coefficient
- brightness per cluster

Decay Time: yields size of object



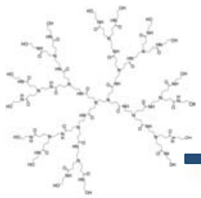
$$G(\tau) = \frac{\langle \delta I(t) \bullet \delta I(t+\tau) \rangle}{\langle I(t) \rangle^2}$$

At τ=0, goes as 1/N



$$\tau_c = \frac{\omega_o^2}{4D}$$

Werner, J. H.et al. *P. N. A. S.* (USA) 11130-11135 (2006)



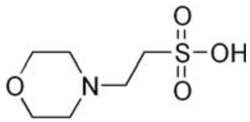
Polyamine dendrimers: template dictates size of cluster

- Size of cluster: ~2.2 nm
- Concentration: ~10 uM
- Fluorescent cluster to dendrimer ratio: 22%
- Brightness per cluster: ~0.85 kHz at 20uW

	QD605	norharmane	carboxy blue
Diameter (nm)	16	0.81	2.2
Diffusion coefficient (um^2/s)	28.81	530	195
Diffusion time tau (us)	1070	52	146

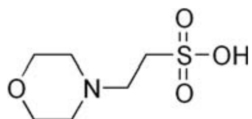
Carboxy blue	Norharmane	QD605 (PEG-amine)	Sengupta'sT4C4T4-Ag
0.85 kHz at 20uW	0.48 kHz at 20uW	60 kHz at 0.5 uW	0.6 kHz at 30 uW

Yeh, H.-C., et. al. "Photophysical characterization of fluorescent metal nanoclusters synthesized using oligonucleotides, proteins, and small molecule ligands," *Proc. of SPIE* 7576, 75760N-75761, (2010)

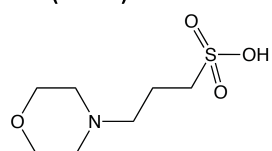


Small molecule templates: morpholine and piperazine

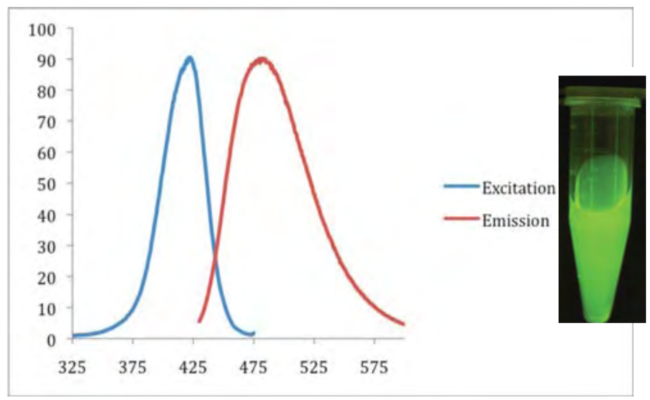
Motivation: small molecule template/small size aggregate; small and tunable ligands for theory; nanoparticles formed in GOODS buffers (Xie Chem. Mater. 2007)

 + $HAuCl_4$ $\xrightarrow[\text{3 days}]{\text{37 °C}}$ Au-nanoclusters

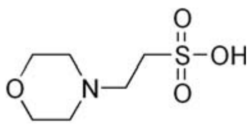
3-(N-morpholino)ethanesulphonic acid
(MES)



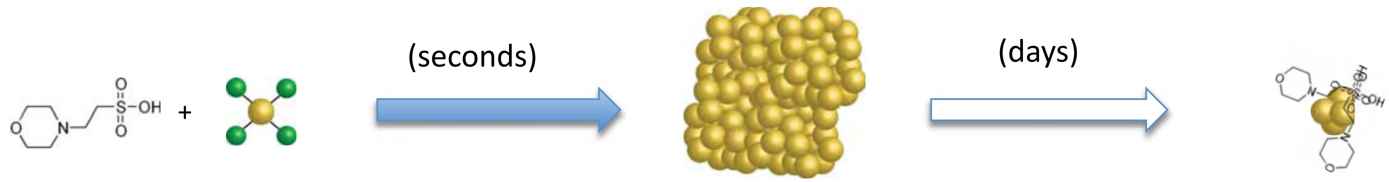
3-(N-morpholino)propanesulphonic acid
(MOPS)



Bao, Y, et. al. "Formation and Stabilization of Fluorescent Au Nanoclusters using Small Molecules," *J. P. C. C.* 114(38), 15879-15882 (2010)



Small molecule templates produce clusters through etching:



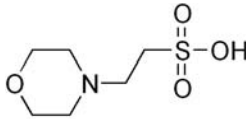
MES

$[AuCl_4]^-$

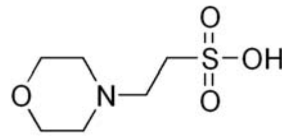
Gold particle
scatters light, has plasmon resonance

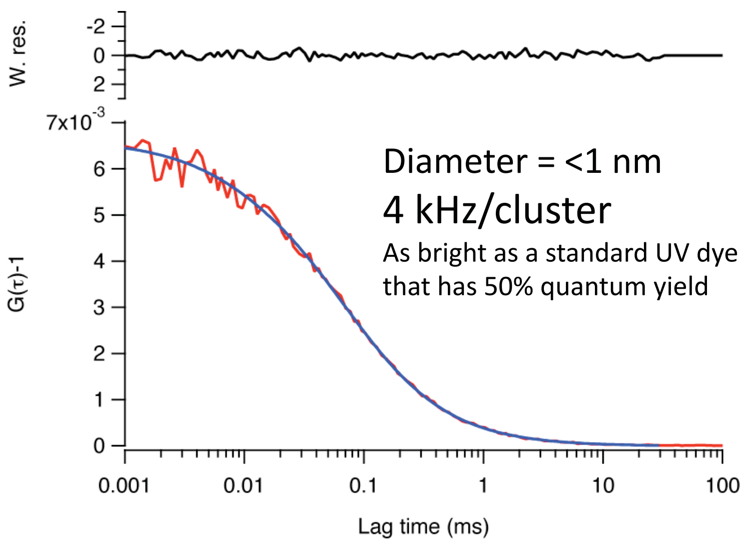
Gold nanocluster
Fluorescent, no plasmon resonance

- Facile synthesis
- Many small molecules produce fluorescence
- Amines seem to be a key functional group

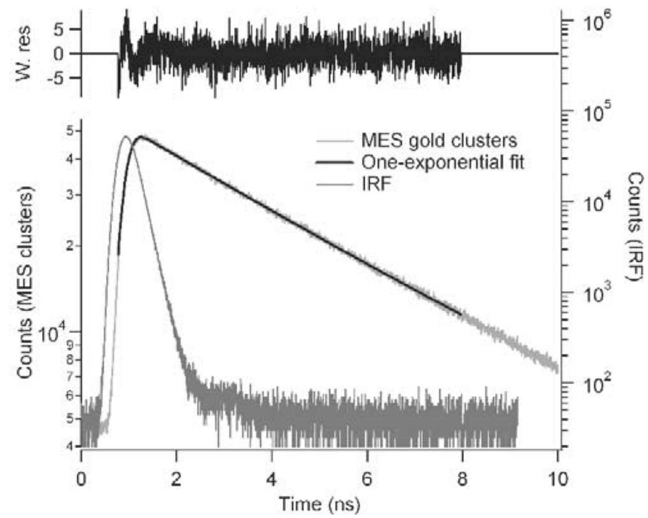


Small molecule templates produce clusters are small and bright:

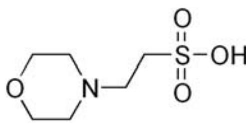




FCS fluorescence correlation spectroscopy

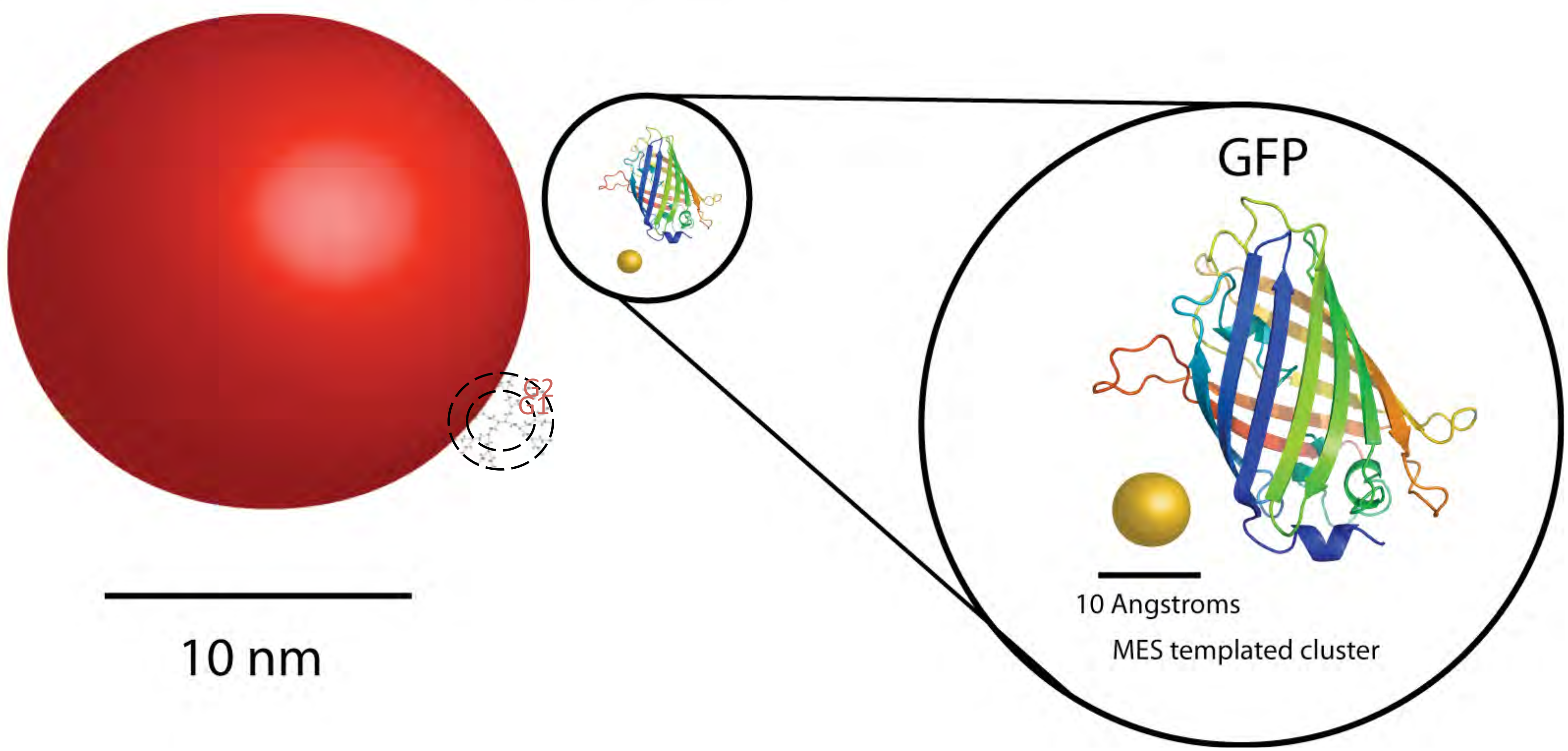


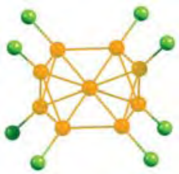
TCSPC time correlated single photon counting



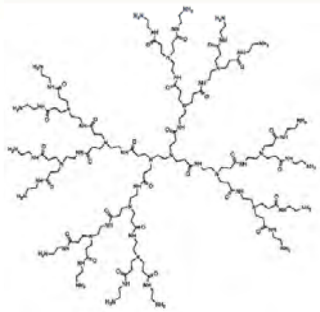
Small molecule templates produce clusters are small and bright:

Quantum Dot (CdSe, ZnS cap, water solubilized)





Mechanistic details are hard to determine from large templates



+ Au^{III} → **?** → **fluorescent species**

Mechanism?

Cluster?

Neutral?

Au clusters → cationic (only known with PR_3, X^-, RS^-)

Au clusters → **neutral** (unknown with ligands)

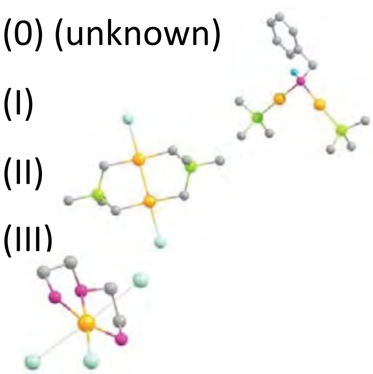
Au clusters ↓ **Au complexes**

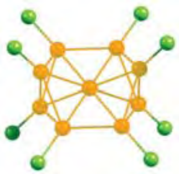
Au complexes → Au(0) (unknown)

Au complexes → Au(I)

Au complexes → Au(II)

Au complexes → Au(III)

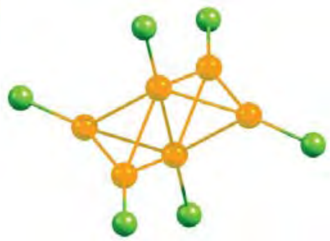




Cationic clusters: defined starting materials

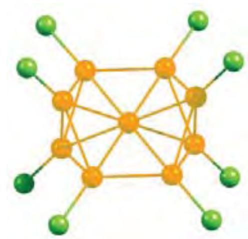
No small stable and *neutral* ligated Au^0 clusters are known,
but bare Au_2, Au_3, and Au_4 were shown to emit in gas phase/Ar matrix

$Au_4(PR_3)_4^{2+}$

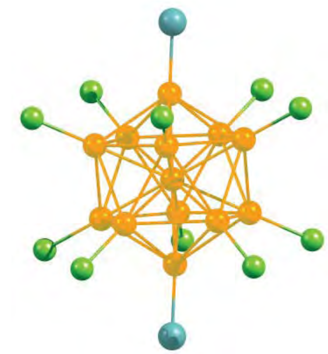


$Au_6(PR_3)_6^{2+}$
$\{Au_6^{2+}\}$

$Au_8(PR_3)_8^{2+}$
$\{Au_8^{2+}\}$



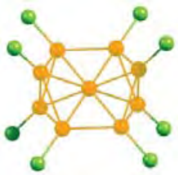
$Au_9(PR_3)_8^{3+}$
$\{Au_9^{3+}\}$

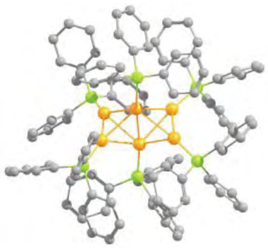


$Au_{13}(PR_3)_{10}Cl_2^{3+}$

❖"Pre-reduced" , with Au ox. state between 0 and 1
❖ None fluorescent

Cationic clusters: defined starting materials

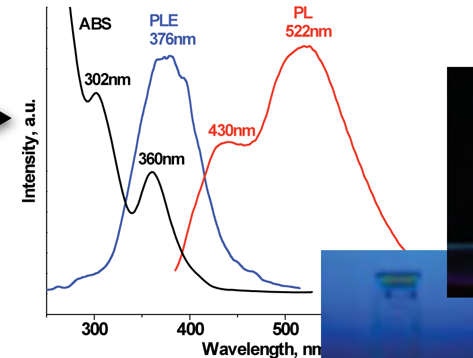


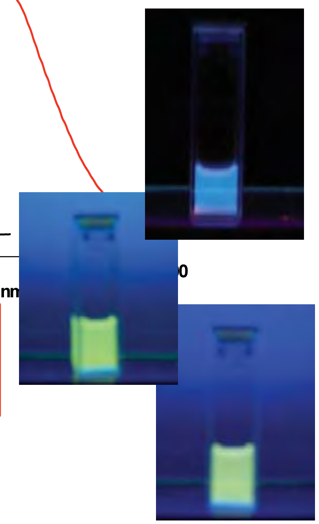


+ $nR\text{-}NH_2$ $\xrightarrow[\text{rm T}]{\text{solvent, 2-3 days}}$ **Au NPs** $\xrightarrow[\text{2-3 days}]{\text{etching}}$

$^{i}PrNH_2$
$OctylNH_2$
G3-PAMAM-NH_2
$^{n}BuNH_2$

Primary amine – fast reaction
Secondary amine – slow reaction
Tertiary amine – no reaction





τ_{rad} = 1-7 ns
bright PL

TLC/HPLC
KCN and HCl quench emission $\longmapsto$ Fluorescence from Au-containing species

Au 4*f* XPS BE < 84eV $\longmapsto$ Neutral gold

no ^{31}P NMR signal from emissive samples $\longmapsto$ No phosphine ligation to gold in emissive species

no emission from amines oxidized by O_2, H_2O_2, electrochemistry $\longmapsto$ Gold is needed for emission

Fluorescence is caused by gold cluster in the presence of amine (weak ligand)

Reaction pathway toward neutral fluorescent gold clusters

1. Amine induced cationic cluster conversion: $Au_9(P)_8^{3+} \longrightarrow Au_8(P)_8^{2+} \longrightarrow Au_6(P)_6^{2+}$ → From UV-Vis

+ Cluster dissociation: $Au_6(P)_6^{2+} \rightleftarrows 2Au_3(P)_3^{+}$ → Follows from ESI-MS and ^{31}P NMR; Supported by DFT calculations

2. Neutral cluster generation: $Au_3(P)_3^{+} + xR' \;\; NH_2 \longrightarrow Au_2(P)_2 + Au(P)(N)^{+}$ → Favorable from DFT calcs

3. Cluster augmentation and breakup and/or amine enrichment:

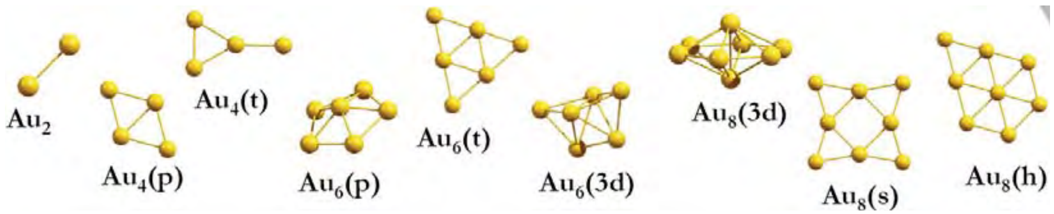
$\{Au_3^{+}\}_P + \{Au_2\}_P \longrightarrow \{Au_5^{+}\}_P \longrightarrow \ldots \longrightarrow$ **larger $\{Au_x^{+}\}_P$ clusters** $\xrightarrow[\text{amine deficiency}]{}$ Au NPs

larger $\{Au_x^{+}\}_P$ clusters $\xrightarrow{\text{amine enrichment}}$ $\boxed{\{Au_2\}_N \rightleftarrows \{Au_4\}_N \rightleftarrows \{Au_6\}_N \ldots}$

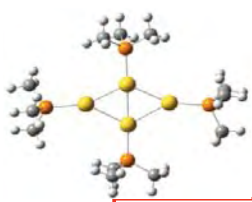
Fluorescent clusters — amine-enriched cluster

> Amine (*or other weak ligand*) is essential to generate fluorescent gold cluster

Theoretical study of small Au clusters: ligand and size effects

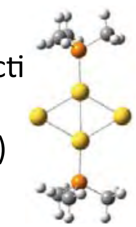


-How does size change electronic and optical properties?
-How does ligand type, geometry, composition change optical properties?
(provide insight on experimental observations)



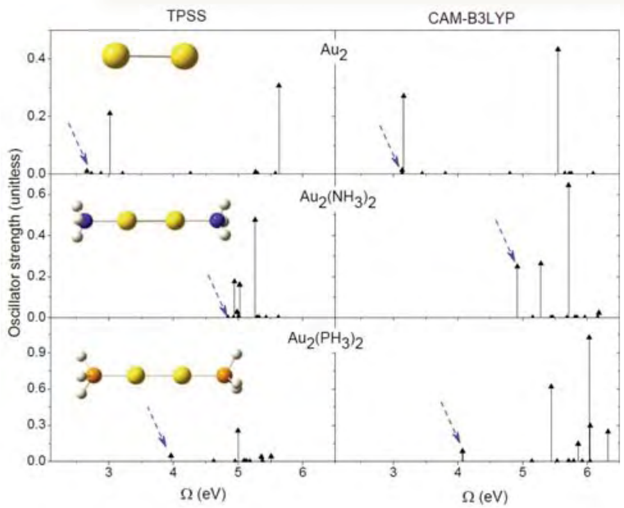
Methods: (TD)DFT approach with LSDA, hybrid GGA and long-range corrected GGA functi
SWVN5, PBEPBE, PBE0, CAM-B3LYP, LC-wPBE

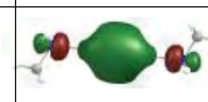
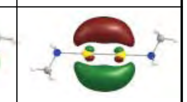
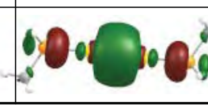
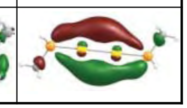
+ ligand influence on excited state structure of neutral gold clusters (Au_2 and Au_4)
+ absorption profile of bare and ligated



- DFT calculation on small gold clusters predict variety of conformations but not the energy scaling for small clusters.
-Thermal fluctuations cause strong conformational freedom.
- Metal-ligand binding energies $NH_3 \approx NMe_3 < PH_3 < PMe_3$
- Metal-ligand binding depends on size and ligand number: $Au_4L_4 < Au_2L_2 < Au_4L_2$

Theoretical study of small Au clusters: ligand and size effects



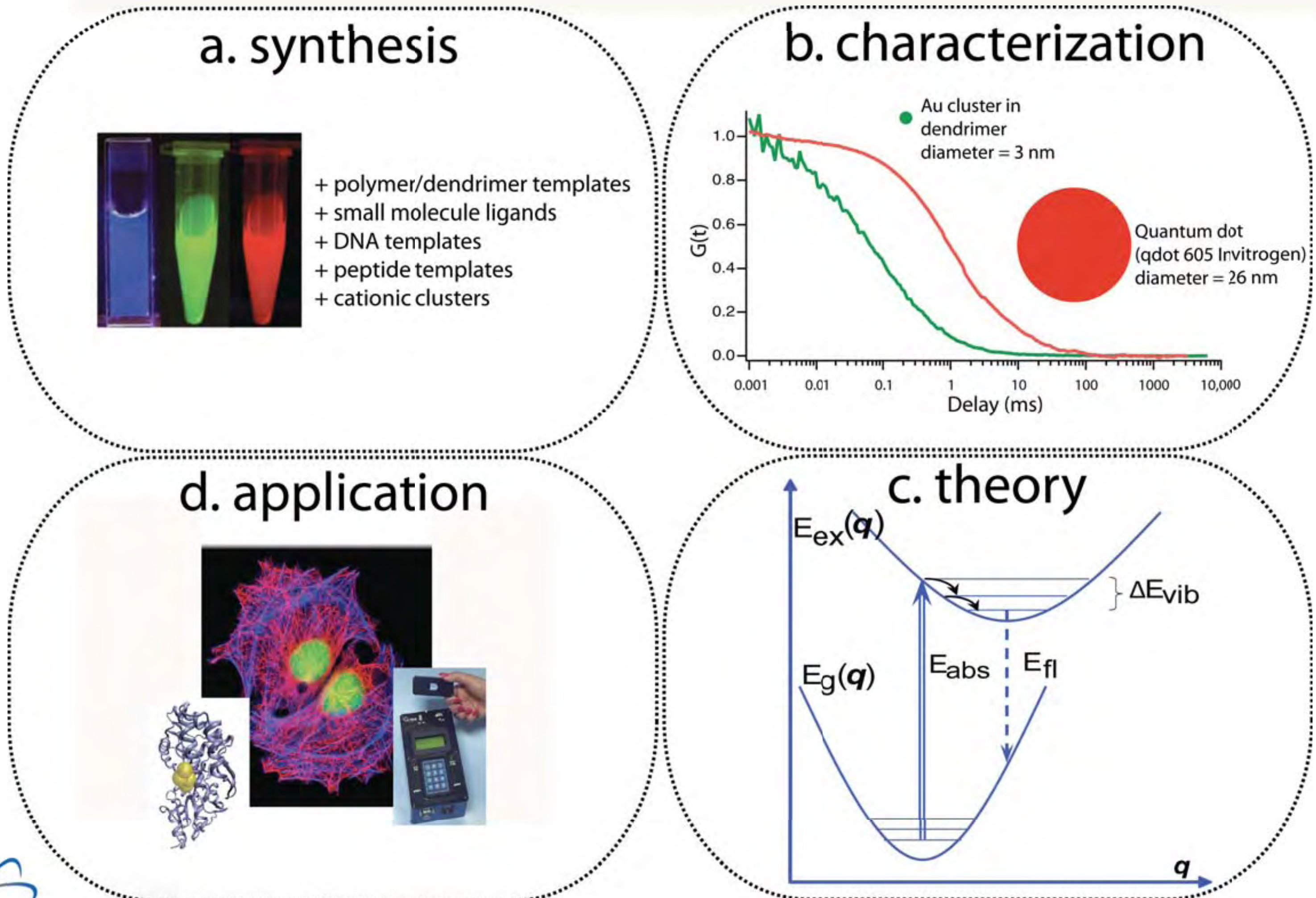
Ligand	f	BE	d(Au-Au)	HNTO	ENTO
CH_3NH_2	0.216	24.4	2.514		
CH_3PH_2	0.059	31.5	2.568		

Size: non-monotonous dependence of optical properties
Ligands: type and number critical
Ligands: modify the excited state structure of clusters by

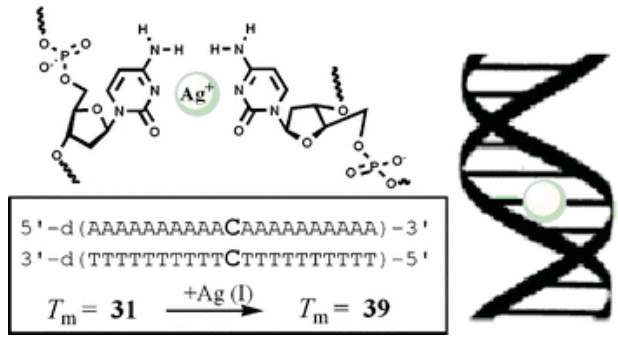
+ *eliminating low-lying optically inactive excited states* and by
+ *controlling the degree of metal-ligand delocalization of an excitation*.

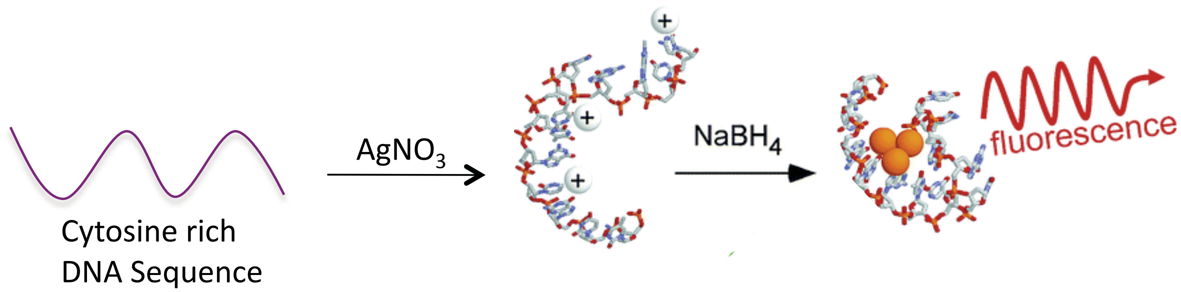
Small Au clusters ligated with *amines will have better fluorescence potential* compared to those ligated with phosphine or thiol ligands, in agreement with preliminary experimental data.

Goel S., et. al. "DFT Study of Ligand Binding to Small Gold Clusters", J. Chem. Phys. Lett., 2010, pp 927-931.
Goel S., et. al. "DFT Study of Optical Transitions in Bare and Ligated Small Gold Clusters", J. Chem. Phys. Lett. (submitted)
Ivanov, S. et. al. "Density Functional Analysis of Geometries and Electronic Structures of Gold-Phosphine Clusters," J. Phys. Chem. C. (accepted 2011)


a. synthesis
+ polymer/dendrimer templates
+ small molecule ligands
+ DNA templates
+ peptide templates
+ cationic clusters
b. characterization
Au cluster in dendrimer diameter = 3 nm
Quantum dot (qdot 605 Invitrogen) diameter = 26 nm
G(t)
1.0
0.8
0.6
0.4
0.2
0.0
0.001
0.01
0.1
1
10
100
1000
10,000
Delay (ms)
c. theory
$E_{ex}(\boldsymbol{q})$
ΔE_{vib}
E_{abs}
E_{fl}
$E_g(\boldsymbol{q})$
$\boldsymbol{q}$
d. application

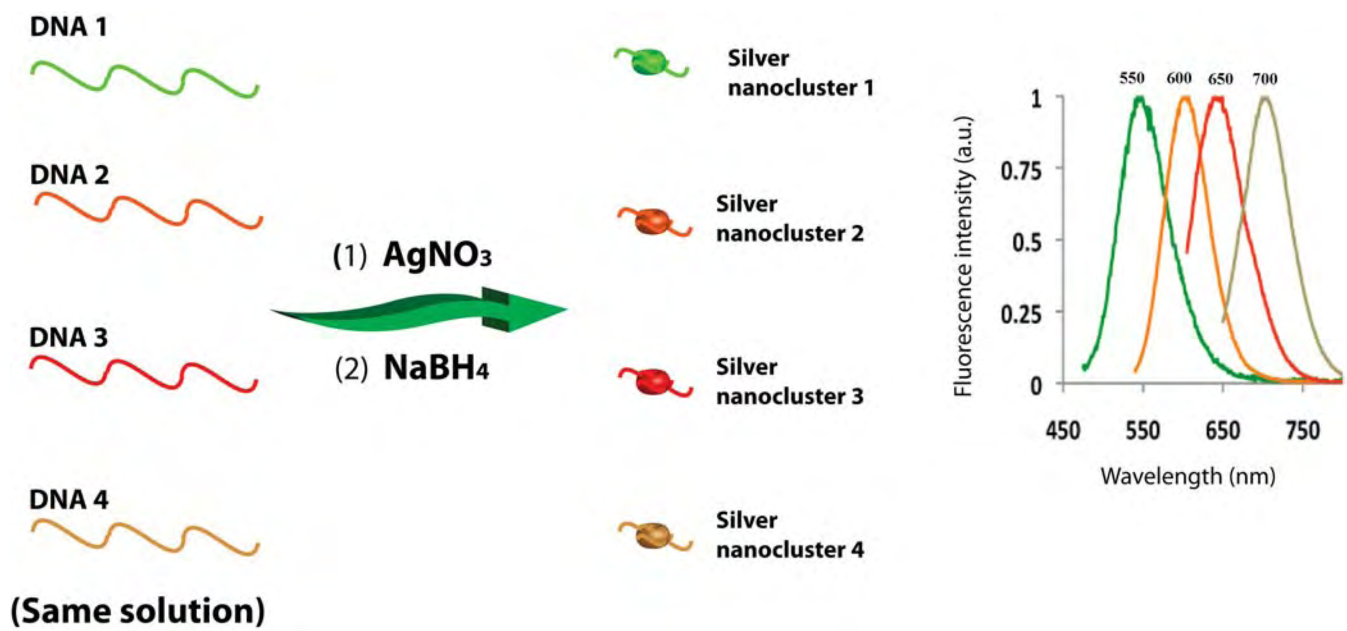
DNA templates:



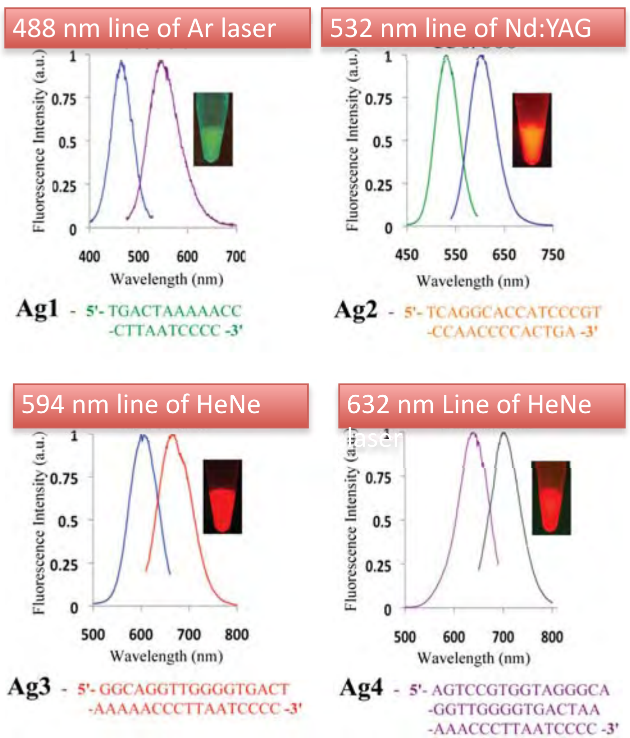


Dickson et al. *J. Am. Chem. Soc.*, 130, 5038-5039, 2008
Petty et al., *J. Am. Chem. Soc.*, 126, 5207-5212, 2004
O'Neil et al. *Journal of Physical Chemistry C* 113, 2009

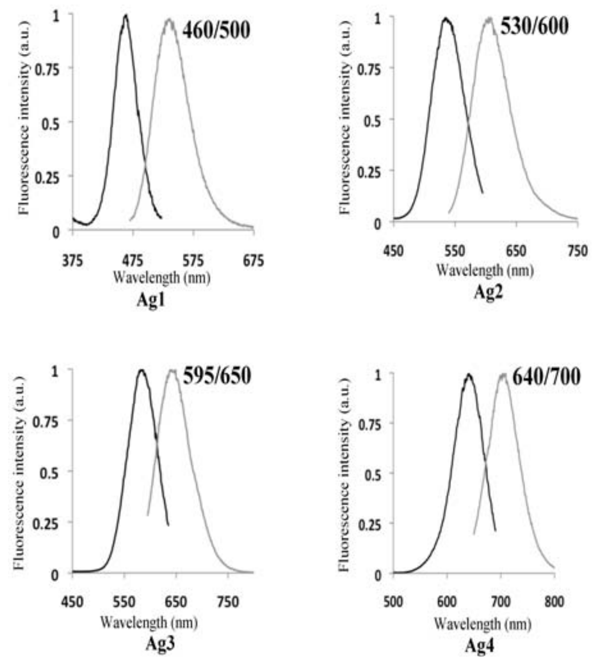
Developing a palette of fluorophores



DNA templates: wavelength tunable Ag-clusters



Phosphate buffer (pH 5.8 -7.5)



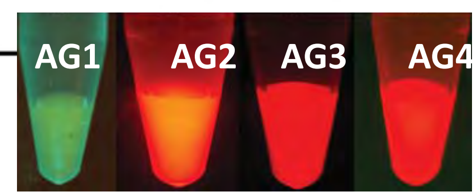
Water

Sequence dependent fluorescence

Sharma, J., et. al. "A complimentary palette of fluorescent silver nanoclusters," *Chem. Commun.* 46, 3280-3282 (2010)

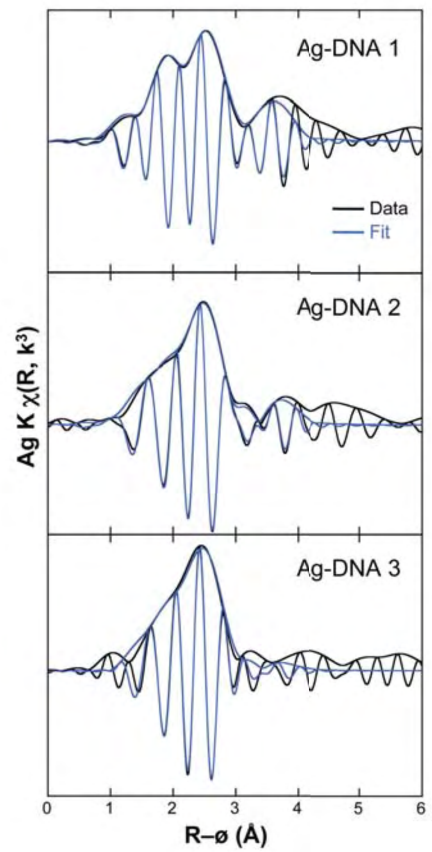
DNA templates: wavelength tunable Ag-clusters

Nanocluster	Emission peak (nm)	Lifetime (ns)	Fractional intensity	Quantum yield
AG1	550	2.5 0.59	38 62	0.002 ± 0. 0002
AG2	600	2.86 0.8	59 41	0.107 ± 0. 014
AG3	650	3.47 1.32	64 36	**0.640 ± 0. 014**
AG4	700	3.6		0.524 ± 0. 034



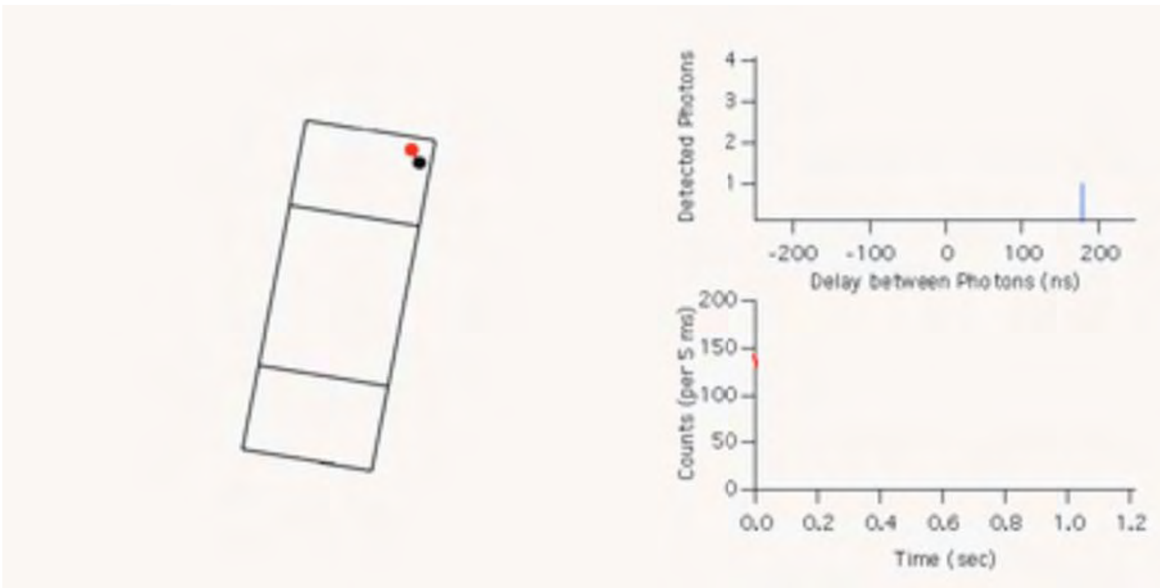
EXAFS of DNA-Ag clusters: Ag-Ag and Ag-DNA interactions

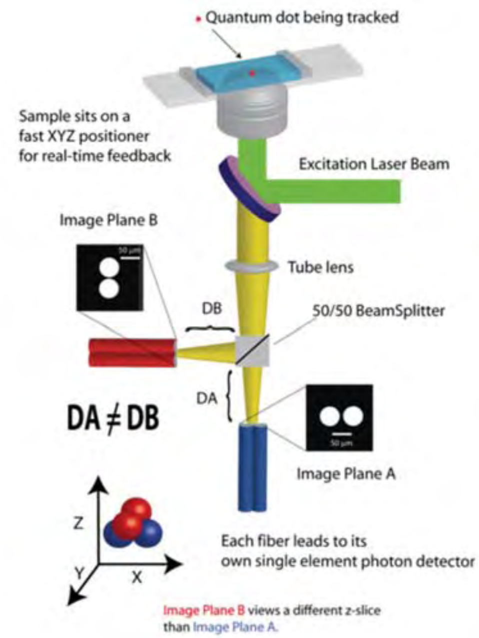
- Presence of Ag-Ag bonds at distances contracted from Ag metal, consistent with the presence of small clusters
- DNA directly ligates to Ag
- Average size of the Ag clusters is Ag_8-Ag_{20}
- Different DNA sequence leads to different Ag speciation/ligand interactions...correlates to SANS

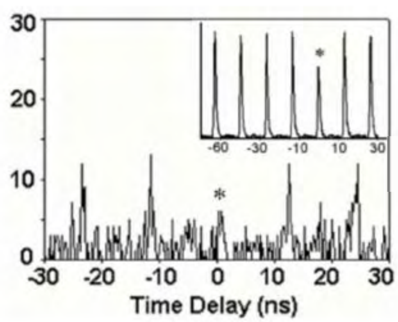


Neidig, M.L. et. al. "Ag K-Edge EXAFS of DNA-Templated Fluorescent Silver Nanoclusters: Insight into the Structural Origins of Emission Tuning by DNA Sequence Variations" *J. Amer. Chem. Soc.* (in review)

3D tracking and fluorescence photon anti-bunching of single molecules

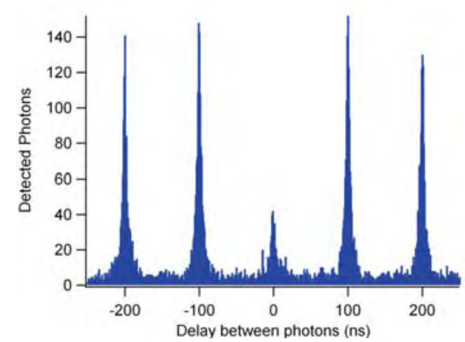






Stuck on a surface. Not very convincing anti-bunching

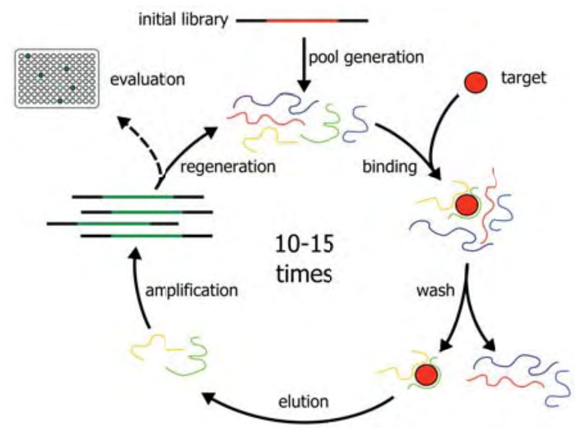
Moving in 3D at biologically relevant transport rates. Clear anti-bunching



"Nanoparticle-Free Single Molecule Anti-Stokes Raman Spectroscopy" Peyser-Capadona, Gonzalez, Lee, Patel, Dickson Phys. Rev. Lett. 94 058301 (2005)

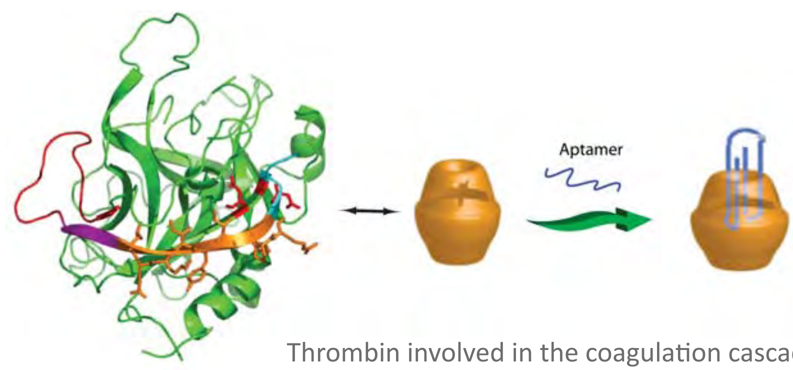
DNA nanoclusters: protein detection

Aptamers = DNA or RNA evolved to bind specific proteins or low-molecular-weight inorganic or organic substrates



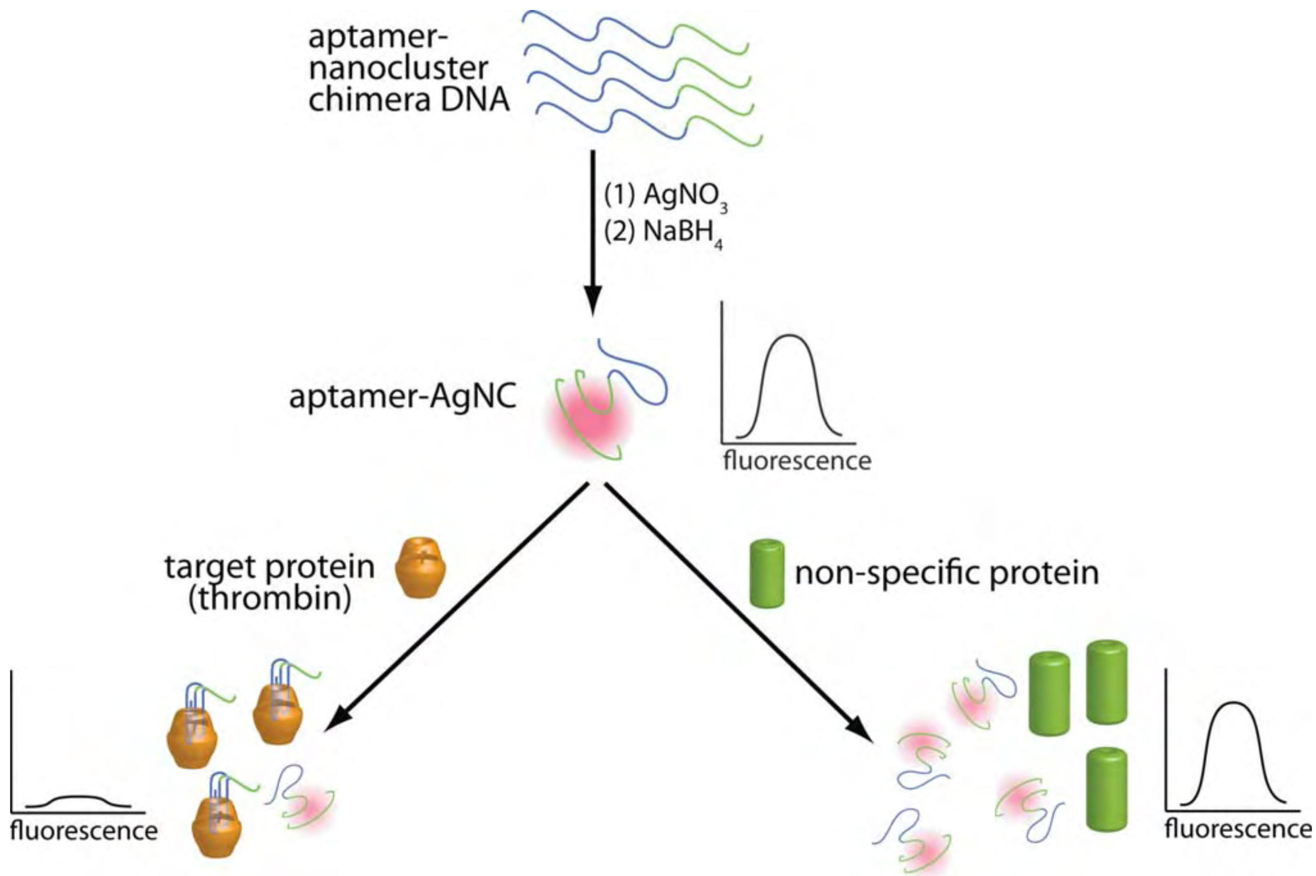
Systematic evolution of ligands by exponential enrichment (SELEX)

Thrombin protein



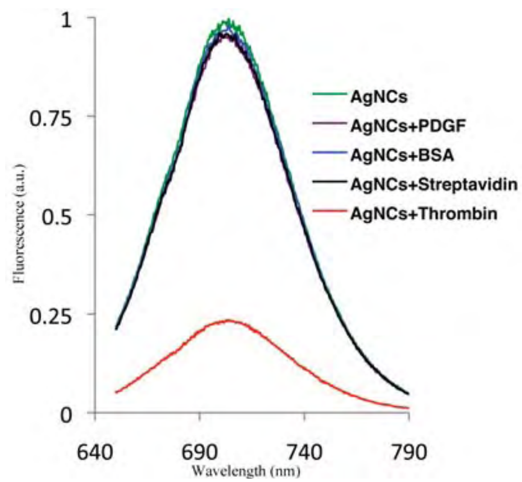
Thrombin involved in the coagulation cascade to convert fibrinogen to insoluble fibrin in physiological conditions or in a pathological thrombus

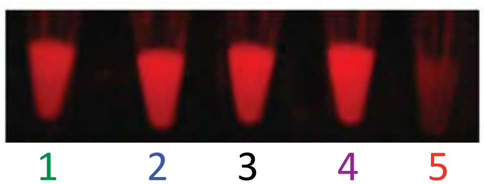
Aptamer-nanocluster chimera detects proteins



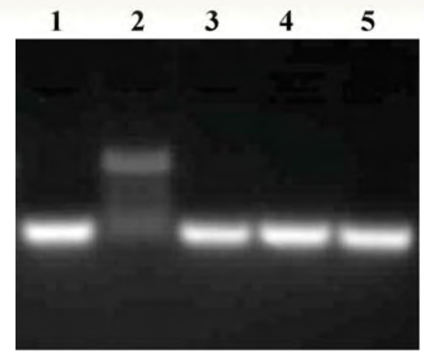
Sharma, J., et. al. "In-situ generation of aptamer templated silver nanoclusters for label-free protein detection," *Chem. Comm.,* DOI: 10.1039/C0CC03711G (2010)

Aptamer-nanocluster: quenching on protein binding

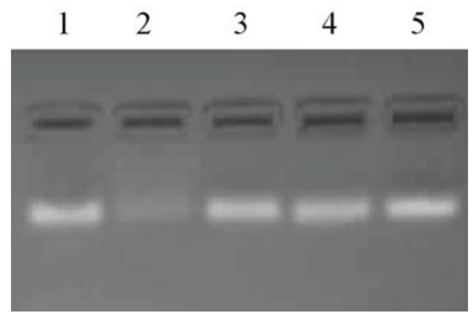




Fluorescence is quenched only on addition of specific protein



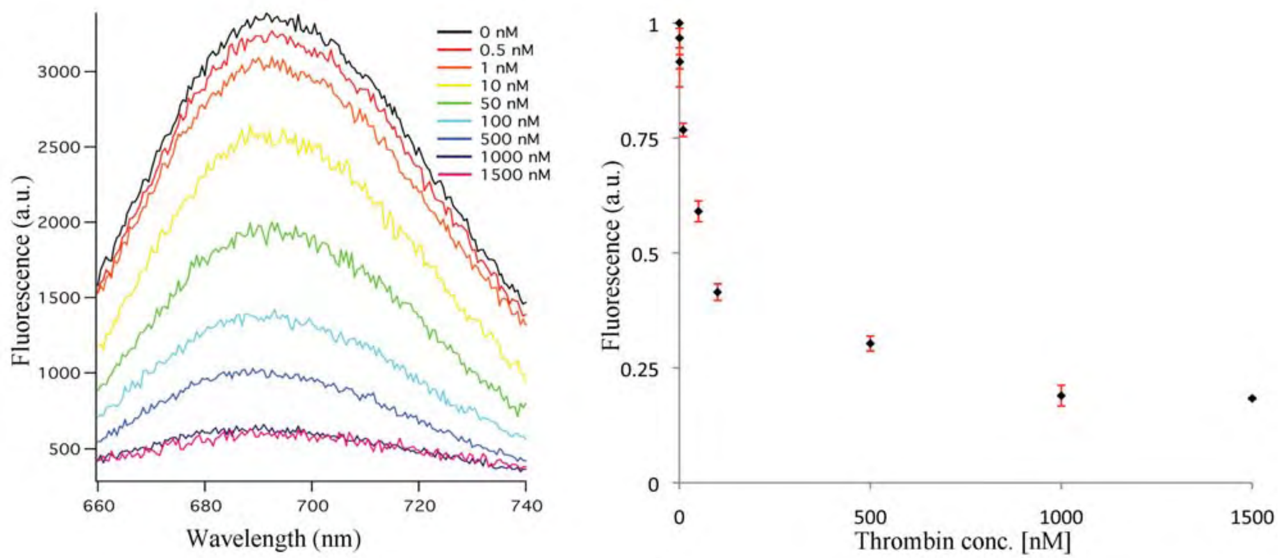
Agarose gel showing specific binding of thrombin (Th) aptamer. Lane 1: AgNC alone, 2: AgNC+Th protein (gel contains ethidium bromide).



Agarose gel showing specific binding of thrombin (Th) aptamer. Lane 1: alone AgNCs, 2: AgNCs+Th protein (bands are visible from AgNC fluorescence).

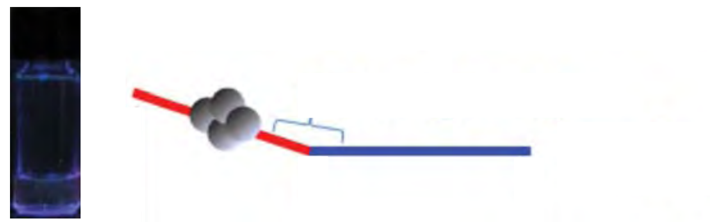
Fluorescence quenching results from protein binding

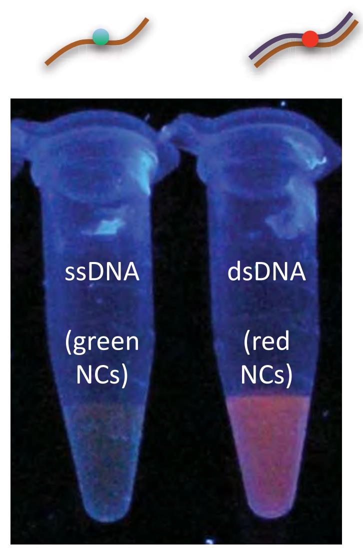
Aptamer-nanocluster: quenching is specific and quantitative



Detection limit of the system is = 1 nM

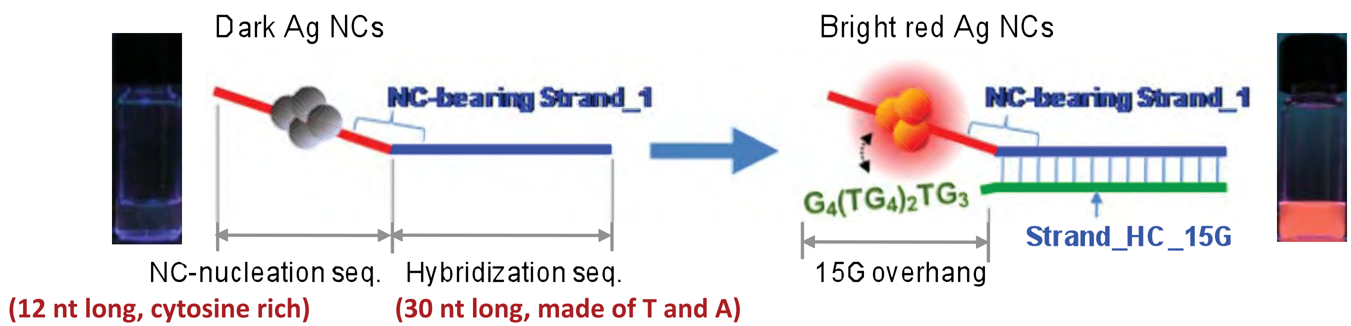
DNA nanoclusters: DNA detection

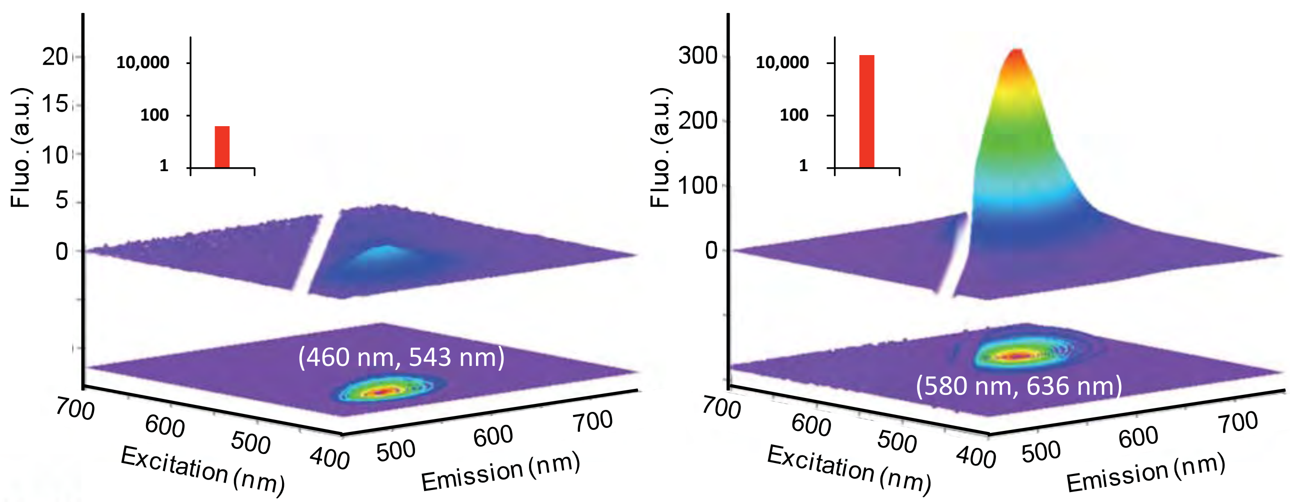




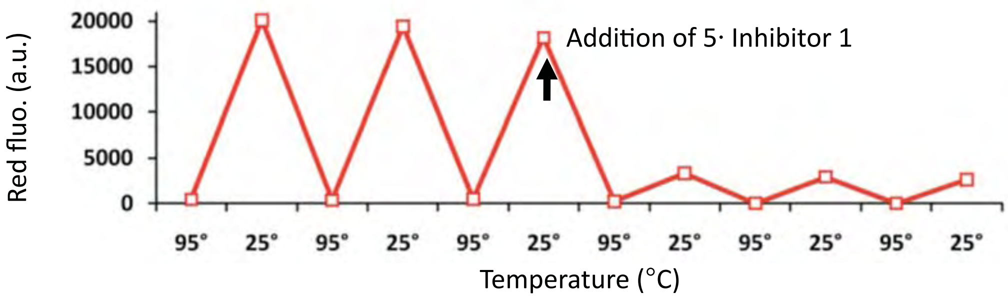
DNA nanoclusters: enhancement on guanine proximity

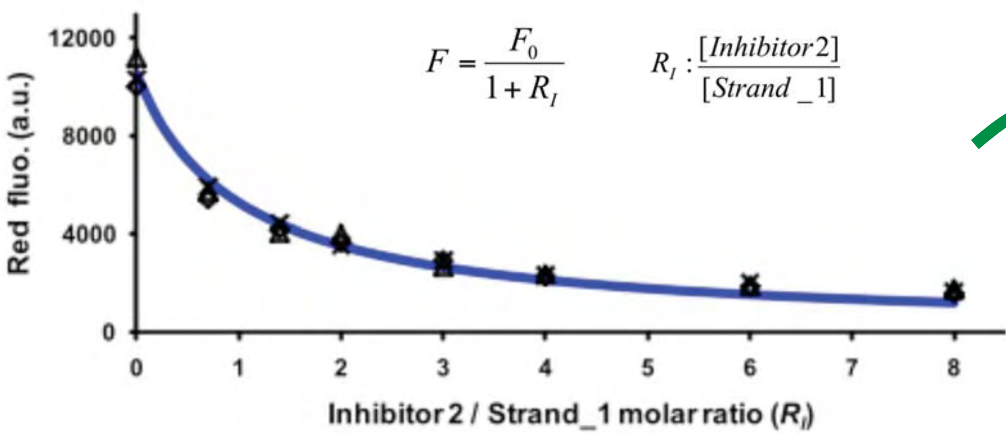
- Guanine base is the key to the observed red fluorescence enhancement.

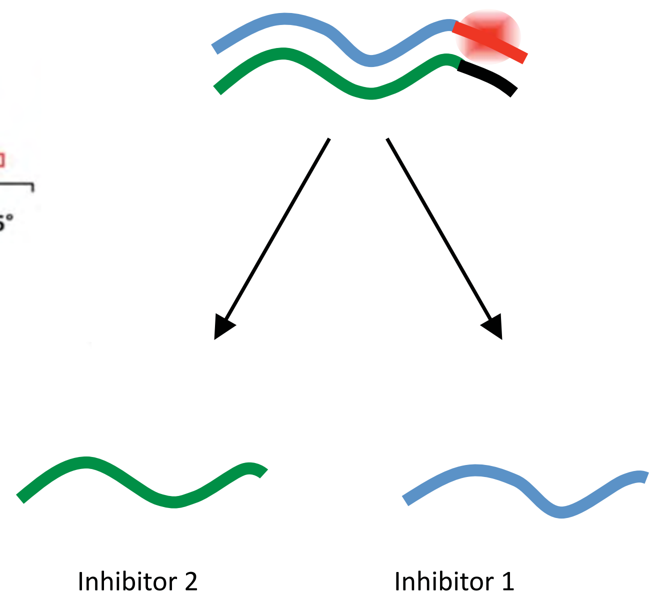




Enhancement is reversible and quantitative

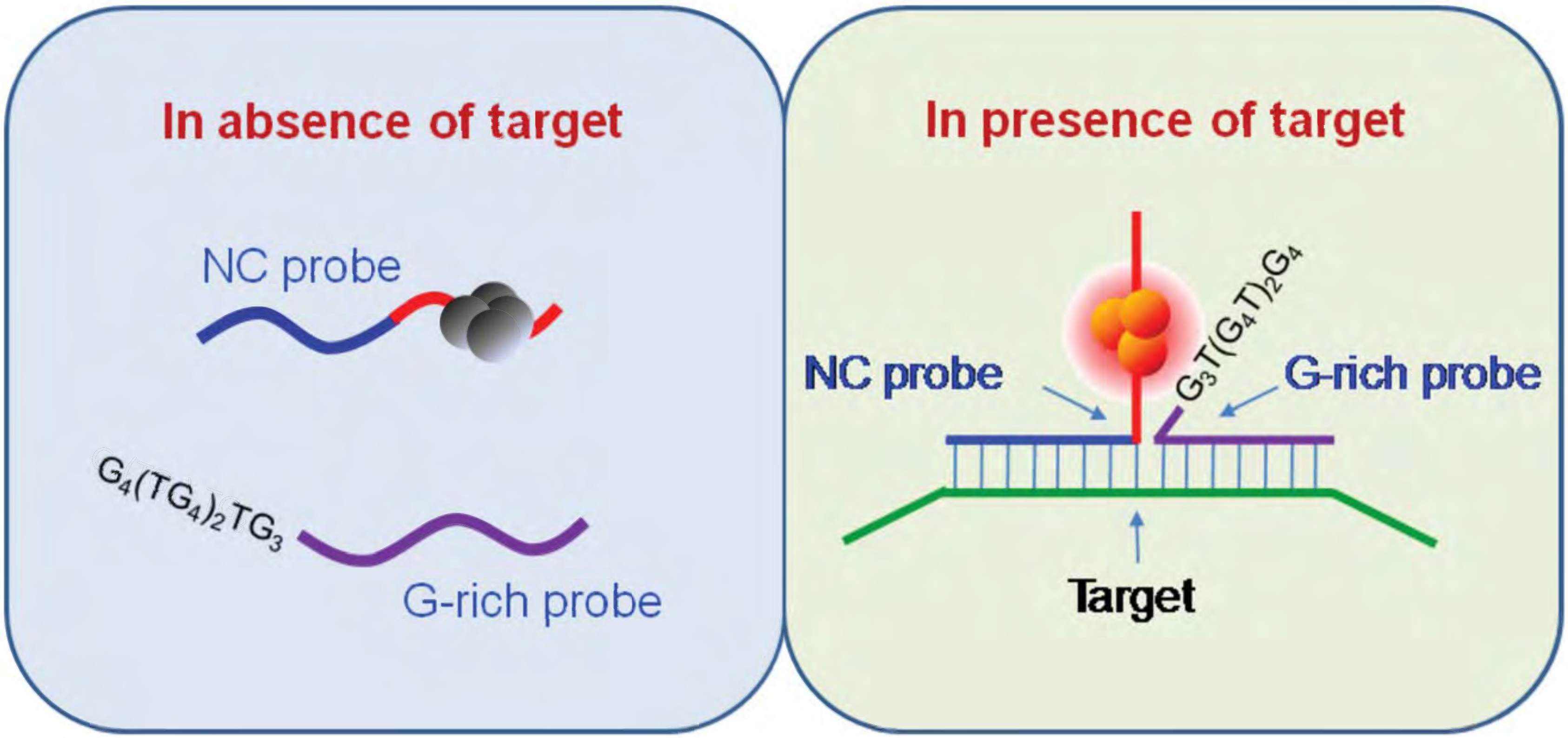






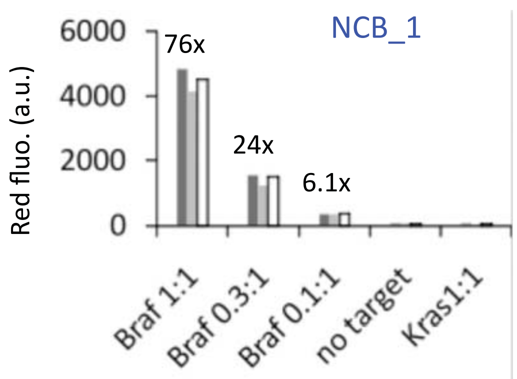
NanoCluster Beacons for DNA detection

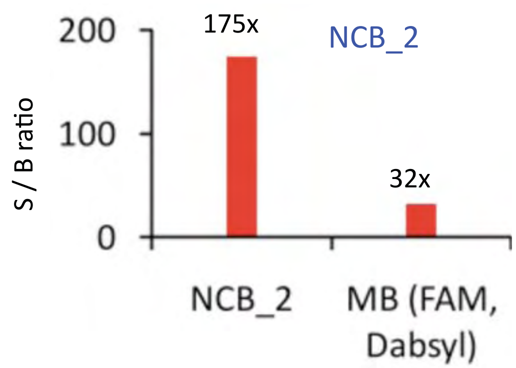
- A new molecular probe based on DNA/Ag NCs.

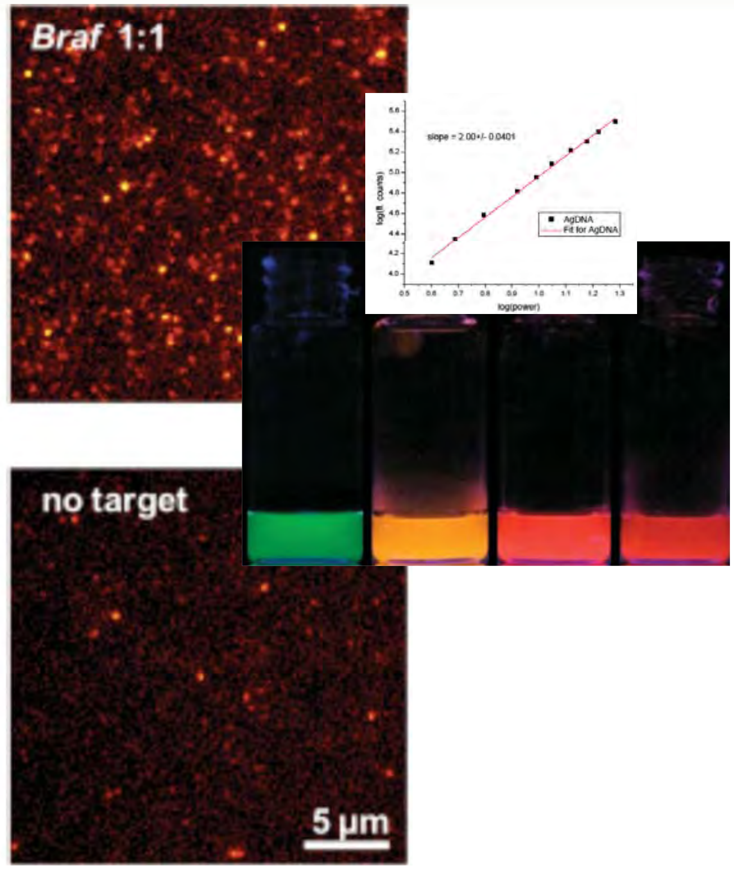


Yeh, H.-C., et. al. "Fluorescence enhancement of DNA-silver nanoclusters from guanine proximity," *Nano Letters* 10(8), 3106-3110 (2010)

NanoCluster Beacons: better than molecular beacon



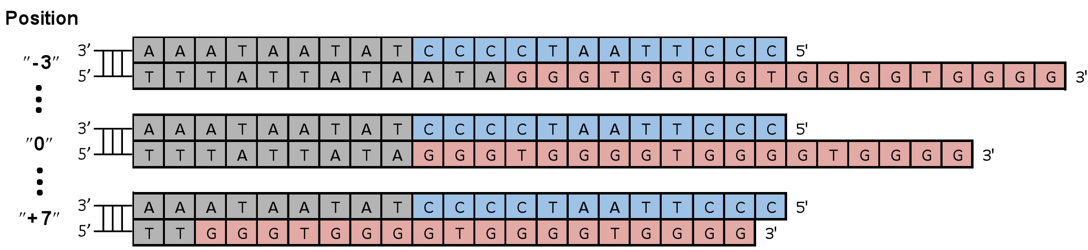




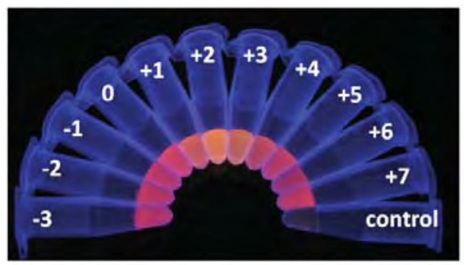
TIRF images of single NCBs acquired by J. Han

- Separation free – no need to do immobilization and wash.
- Excellent S/B ratio – good for single-molecule-based digital assays.
- Two-photon acitive

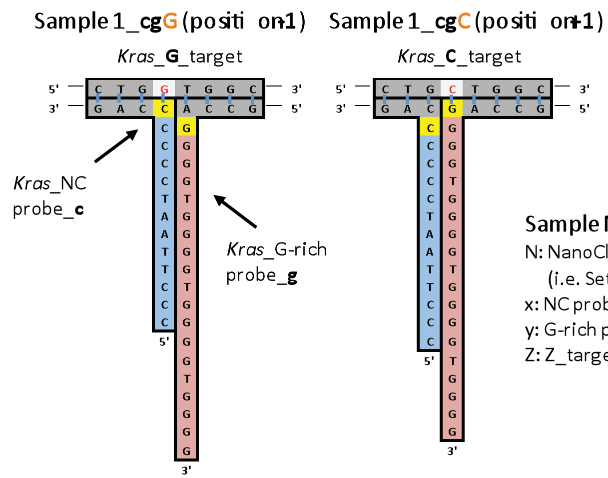
NanoCluster Beacons: fluorescence changes based on environement



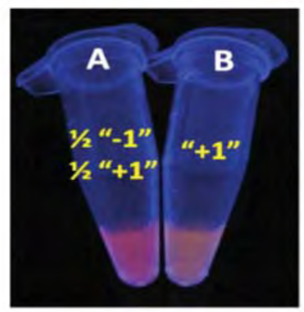
Upper strand: a common NC-bearing strand. Lower strand: individual G-rich strand.
Gray: Hybridization sequences. Blue: NC-nucleation sequence. Red: Enhancer sequence.



Chameleon NanoCluster Beacons: detect SNPs

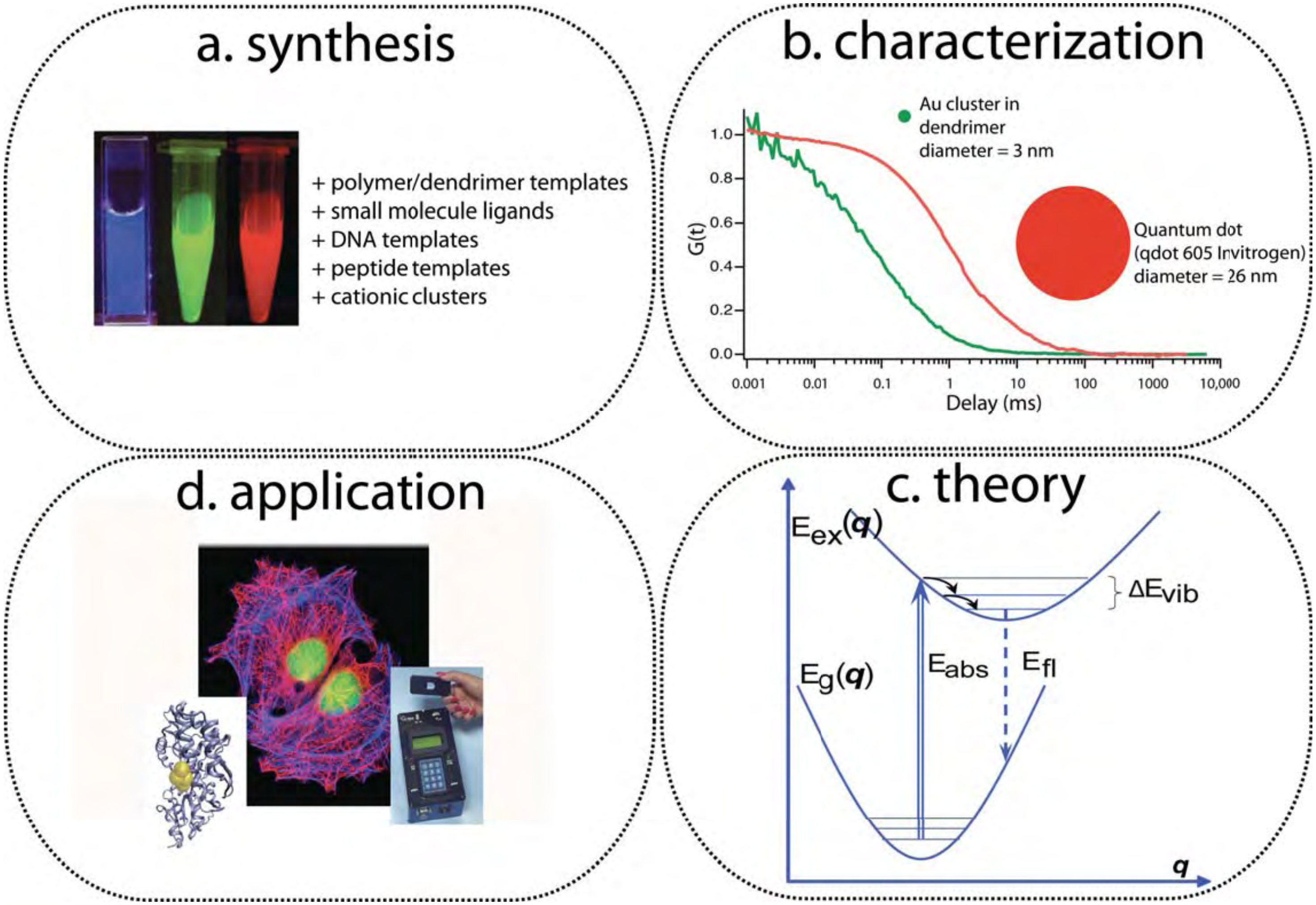


Sample N_xyZ
N: NanoCluster Beacon number
(i.e. Set number)
x: NC probe_x
y: G-rich probe_y
Z: Z_target



- Separation free – no need to do immobilization and wash.
- No heat, no enzymes, more efficient than current methods

Yeh, H.-C. et. al. "Detection of Single-Nucleotide Variations Using Chameleon NanoCluster Beacons"
Nature Biotechnology (submitted)


a. synthesis
+ polymer/dendrimer templates
+ small molecule ligands
+ DNA templates
+ peptide templates
+ cationic clusters
b. characterization
Au cluster in
dendrimer
diameter = 3 nm
Quantum dot
(qdot 605 Invitrogen)
diameter = 26 nm
G(t)
1.0
0.8
0.6
0.4
0.2
0.0
0.001
0.01
0.1
1
10
100
1000
10,000
Delay (ms)
d. application
c. theory
$E_{ex}(\boldsymbol{q})$
ΔE_{vib}
$E_g(\boldsymbol{q})$
E_{abs}
E_{fl}
$\boldsymbol{q}$

Predictive Design of Noble Metal Nanoclusters

Papers/Patents:

-Goel S., et. al. "DFT Study of Ligand Binding to Small Gold Clusters", J. Chem. Phys. Lett., 2010, pp 927-931.
-Goel S., et. al. "DFT Study of Optical Transitions in Bare and Ligated Small Gold Clusters", J. Chem. Phys. Lett. (submitted)
-Ivanov, S. et. al. "Density Functional Analysis of Geometries and Electronic Structures of Gold-Phosphine Clusters," J. Phys. Chem. C. (accepted 2011)
-Yeh, H.-C. et. al. "Detection of Single-Nucleotide Variations Using Chameleon NanoCluster Beacons" Nature Biotechnology (submitted)
-Sharma, J., et. al. "In-situ generation of aptamer templated silver nanoclusters for label-free protein detection," *Chem. Comm.,* DOI: 10.1039/C0CC03711G (2010)
-Yeh, H.-C., et. al. "Fluorescence enhancement of DNA-silver nanoclusters from guanine proximity," *Nano Letters* 10(8), 3106-3110 *(*2010)
-Sharma*, J., et. al. "A complimentary palette of fluorescent silver nanoclusters," *Chem. Commun.* 46, 3280-3282 (2010)
-Bao, Y, et. al. "Formation and Stabilization of Fluorescent Au Nanoclusters using Small Molecules," *J. P. C. C.* 114(38), 15879-15882 (2010)
-Yeh, H.-C., et. al. "Photophysical characterization of fluorescent metal nanoclusters synthesized using oligonucleotides, proteins, and small molecule ligands," *Proc. of SPIE* 7576, 75760N-75761, (2010)
-Neidig, M.L. et. al. "Ag K-Edge EXAFS of DNA-Templated Fluorescent Silver Nanoclusters: Insight into the Structural Origins of Emission Tuning by DNA Sequence Variations" *J. Amer. Chem. Soc.* (submitted)
-Fluorescence-enhancement of DNA-silver nanoclusters from guanine proximity. H.-C, Yeh, J. Sharma, J.S. Martinez, J.H. Werner S-18,917, 2010
-Synthesis of fluorescent metal nanoclusters. J.S. Martinez, R. B. Dyer, D. M. Vu, C. Zhong, Y. Bao. US Patent US7,914,588 B2
-Wavelength tuned silver nanoclusters and aptamer-nanocluster chimera for protein detection, JS Martinez, J. Sharma, JH Werner, HC Yeh Disclosure 10-171

Transition:

BES – New Direction for Renewal
DHS – Seed funding for demonstration and project development

Thank you

Jim Werner; Sergei Ivanov; Andy Shreve; Andrei Piryatinski; Dung Vu; Sergei Tretiak; Ryszard Michalczyk; Tim Yeh; Jaswinder Sharma; Indika Arachchige; Mike Neidig; Kirill Velizhanin; Satyender Goel

discover LDRD

Los Alamos research on materials in radiation environments

J.S. Shlachter (T-DO)

The broad discipline of materials research in radiation environments at Los Alamos contributes to core mission at the Laboratory in energy security, global security, and reliable nuclear deterrence. Research in this area is conducted in several divisions spanning theory, computation, and experiment, and drawing on researchers with physics, chemistry, and material science backgrounds. For some work, the emphasis is on radiation effects in the materials themselves; radiation environments also allow for materials modification and materials analysis. The flagship facility at Los Alamos, the Los Alamos Neutron Science Center or LANSCE, is a major component of both our current and future strategy for materials research in radiation environments through the Ion Production Facility, the Weapons Neutron Research Facility, and the planned Materials Test Station that will be the basis for the Fission Fusion materials Facility in MaRIE. The Center for Materials at Irradiation and Mechanical Extremes (CMIME) and the Ion Beam Materials Laboratory are both significant efforts at Los Alamos that are assisting in the shift from an observation and validation paradigm to one of prediction and control of materials behavior. This talk will highlight research accomplishments on materials in radiation environments, provide some metrics on Los Alamos work relative to that of our peers, and introduce the subsequent talks and posters for this theme area.

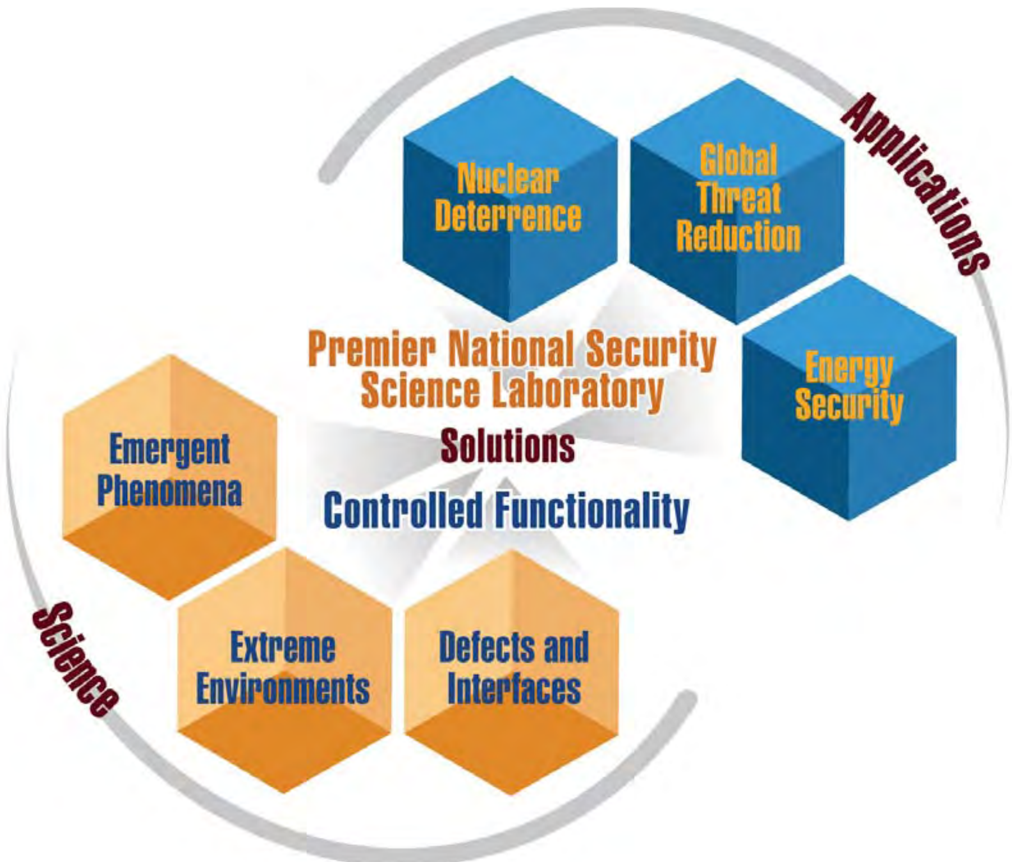
Overview: LANL research on materials in radiation environments

Jack Shlachter

Deputy Division Leader

Theoretical Division

Research on materials in radiation environments contributes to solutions in all LANL applications areas

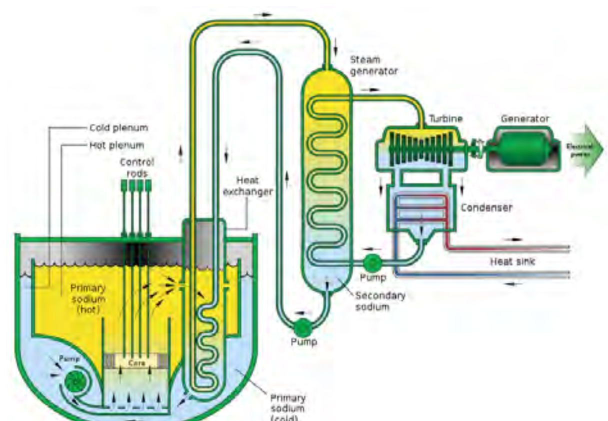


Materials in radiation environments supports *all* major Laboratory missions

Reliable Nuclear Deterrence

Global Security

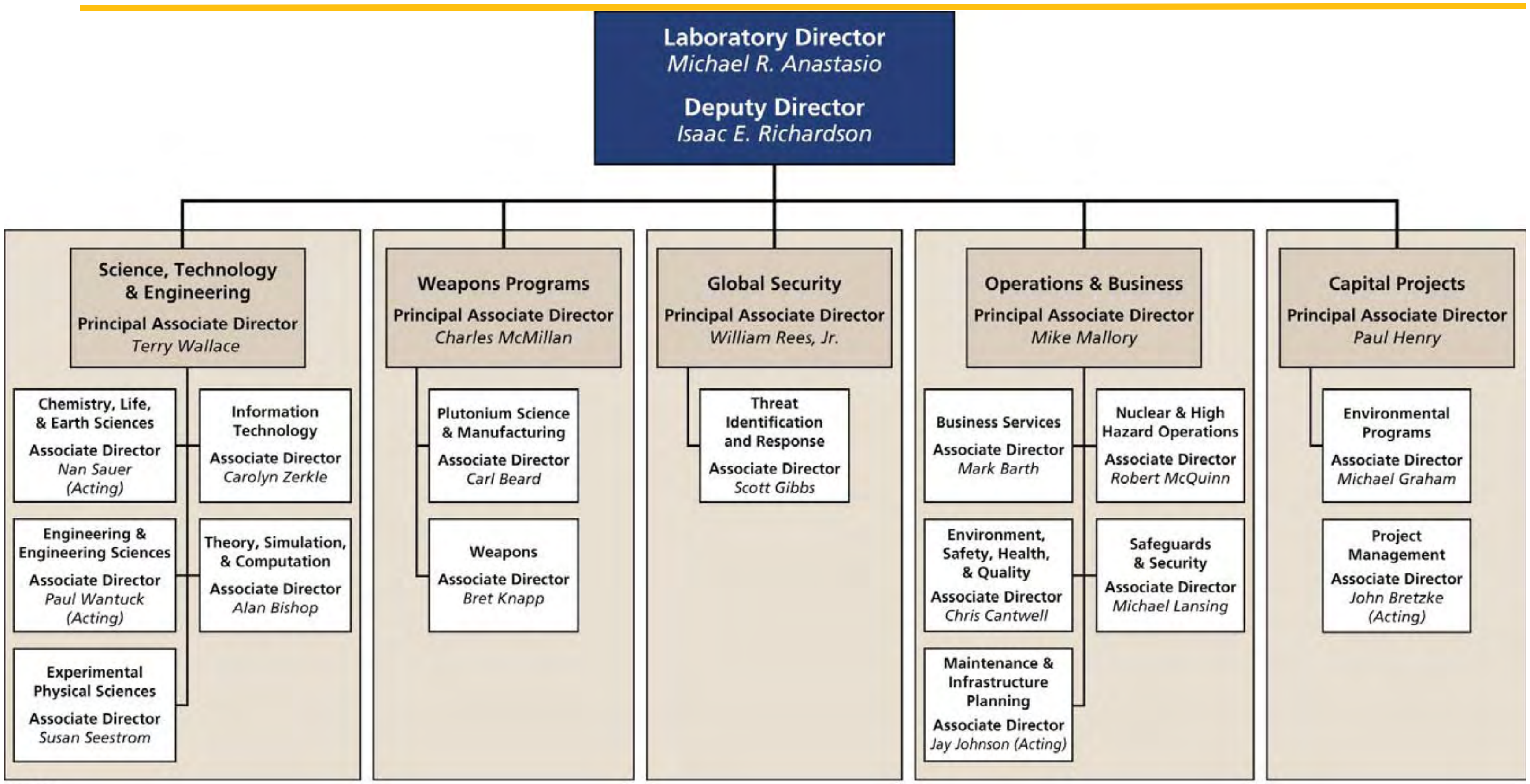
Energy Security



Research on materials in radiation environments is conducted in several Divisions and Centers

- Materials Physics and Applications (MPA)
- Materials Science and Technology (MST)
- Los Alamos Neutron Scattering Center (LANSCE)
- Physics (P)
- Theoretical (T)
- Science Program Office - Civilian Nuclear Programs (SPO-CNP)
- International Space and Response (ISR)
- Nuclear Nonproliferation (N)
- Nuclear Component Operations (NCO)

Research on materials in radiation environments is conducted in several Divisions and Centers



04/05/11

Materials in radiation environments are ubiquitous at Los Alamos

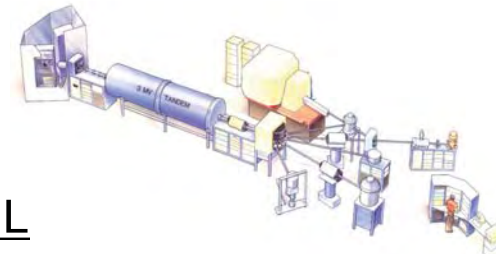
- **Civilian Nuclear Programs**
  - Office of Nuclear Energy (largely Fuel Cycle R&D
  - Earth and Environmental Projects (largely Waste Isolation Pilot Plant)
  - Advanced Simulation and Computation for Environmental Management
- **Weapons aging and certification**
- **New nuclear fuels and/or targets that can safely burn long lived waste products**
- **Radiation effects in semiconductor electronic devices**
- **Nuclear waste characterization**
- **Nuclear data measurements for modeling and simulation of nuclear fuel cycle**
- **Isotopes production programs**
- **Ultra-high efficiency photovoltaics**
- **Nuclear Material detection DNDO**
- **Next-generation nuclear material accounting & detection technologies**

…

Work in radiation environments is synergistic with other materials research

CMIME
Center for Materials at Irradiation and Mechanical Extremes

- Behavior of materials subject to extreme radiation dose
- Behavior of materials subject to extreme mechanical stress



- MTS
- F^3
- WNR
- IPF

IBML

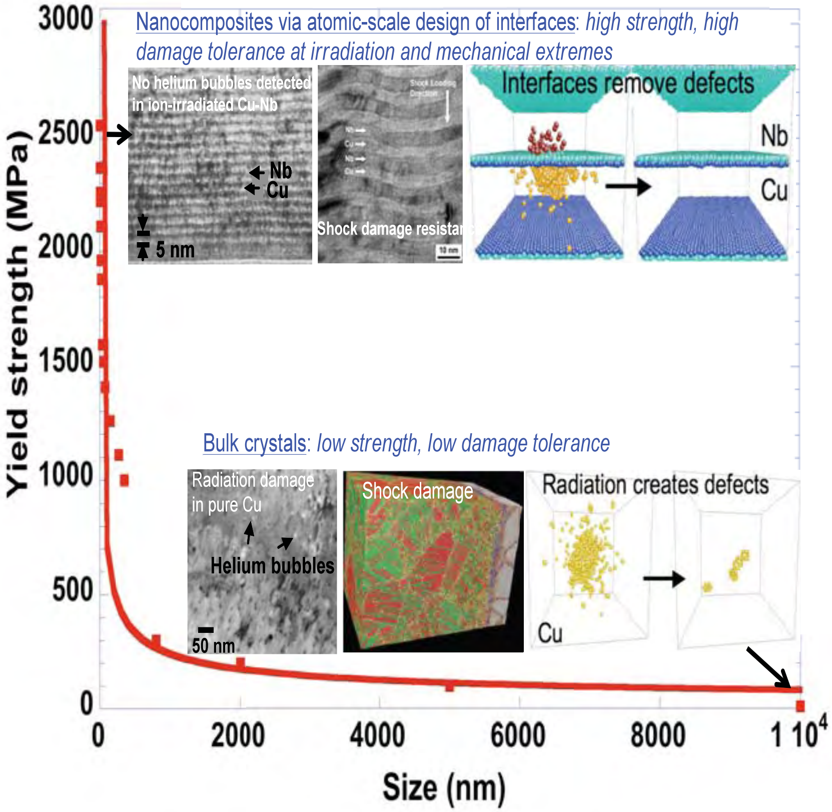
- Materials characterization with **Ion Beam Analysis** techniques
- Materials modification and synthesis through **Ion Implantation**
- **Radiation Damage Effects** in materials by ion bombardment

Fission and fusion reactors

- Materials development and testing for fast reactor core materials
- Fusion first wall material development

Design of interfaces → tailored performance at extremes of irradiation doses and mechanical stresses

CMIME Center for Materials at Irradiation and Mechanical Extremes



CMIME publications

Xian-Ming Bai, Arthur F. Voter, Richard G. Hoagland, Michael A. Nastasi, and Blas P. Uberuaga, "Efficient Annealing of Radiation Damage near Grain Boundaries via Interstitial Emission," *Science*, **327,** 1631 – 1634 (2010).

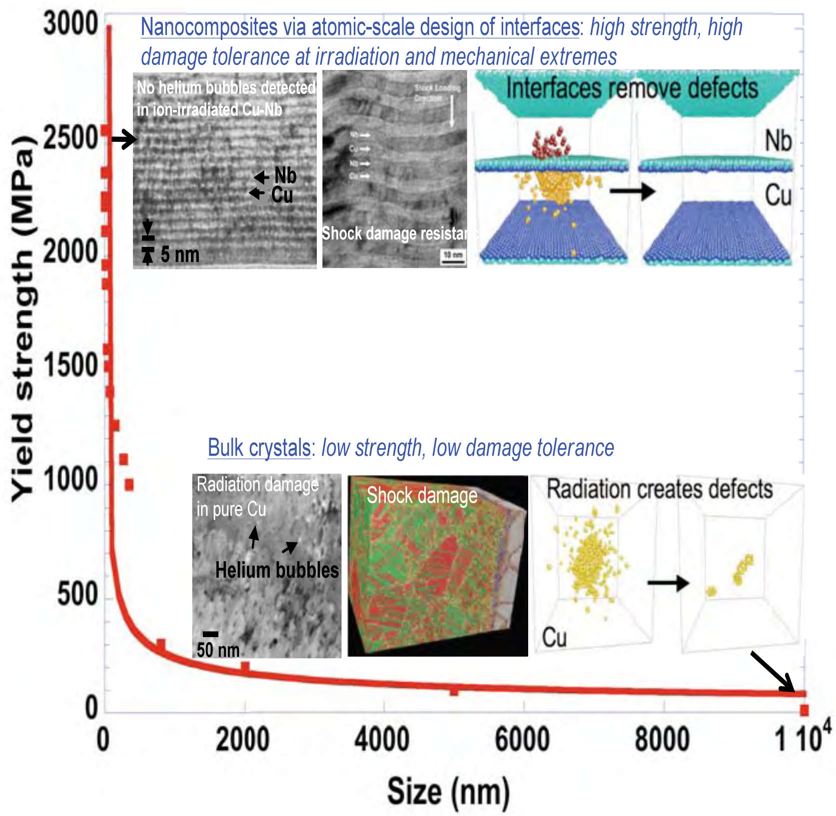
F. Cao, I. J. Beyerlein, F. L. Addessio, B. H. Sencer, C. P. Trujillo, E. K. Cerreta, and G. T. Gray III, "Orientation dependence of shock-induced twinning and substructures in a copper bicrystal," *Acta Materialia* **58**, 549-559 (2010).

S.-N. Luo, T. C. Germann, T. G. Desai, D. L. Tonks, B. P. Uberuaga, and Q. An, "Anisotropic shock response of columnar nanocrystalline Cu," *J. Appl. Phys.* **108**, 093526 (2010).

M. J. Demkowicz, D. Bhattacharyya, I. Usov, Y. Q. Wang, M. Nastasi, and A. Misra, "The effect of excess atomic volume on He bubble formation at fcc-bcc interfaces," *Appl. Phys. Lett*. **97,** 161903 (2010).

Q. M. Wei; Y. Q. Wang; M. Nastasi; A. Misra, "Nucleation and growth of bubbles in He ion-implanted V/Ag multilayers," *Phil. Mag.* **91,** 553 (2011)

Design of interfaces → tailored performance at extremes of irradiation doses and mechanical stresses



Predecessor publications

Interface structure and radiation damage resistance in Cu-Nb multilayer nanocomposites
Demkowicz, MJ ; Hoagland, RG ; Hirth, JP
PHYSICAL REVIEW LETTERS (APR 4 2008) Vol.100, iss.13 136102 (Cited: 24)

The radiation damage tolerance of ultra-high strength nanolayered composites
Misra, A ; Demkowicz, MJ ; Zhang, X ; Hoagland, RG
JOM (SEP 2007) Vol.59, iss.9, p.62-65 (Cited: 23)

Influence of interfaces on the storage of ion-implanted He in multilayered metallic composites
Hochbauer, T ; Misra, A ; Hattar, K ; Hoagland, RG
JOURNAL OF APPLIED PHYSICS (DEC 15 2005) Vol.98, iss.12 123516 (Cited: 23)

Arrest of He bubble growth in Cu-Nb multilayer nanocomposites
Hattar, K. ; Demkowicz, M.J. ; Misra, A. ; Robertson, I.M. ; Hoagland, R.G.
Scripta Materialia (April 2008) vol.58, no.7, p.541-4 (Cited: 8)

Complementary approaches to irradiate materials for testing are invaluable

Reactor and spallation source irradiations

- Up to 25 dpa/year possible
- Bulk sample irradiations in the true environment
- Materials will be radioactive
- Reactor campaigns are time consuming, costly, require long term planning, use precious space

Ion beam irradiations

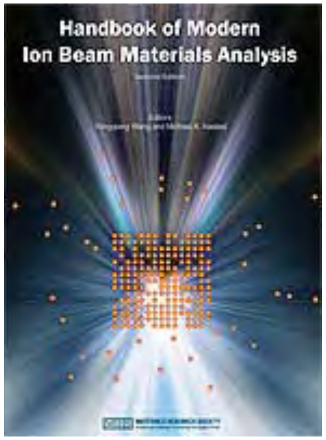
- Attempt to mimic reactor irradiation conditions
- Material will be less radioactive
- Dose rates of several dpa/day are possible
- Cost effective
- Limited penetration depth of ions in matter

IBML has generated over 800 publications in its 25 year history

NEC 3 MV Tandem Ion Accelerator

Danfysik High Current Research Ion Implanter

Supporting programs in BES, NE, DP, NN, WFO, and LDRD



Some recent publications

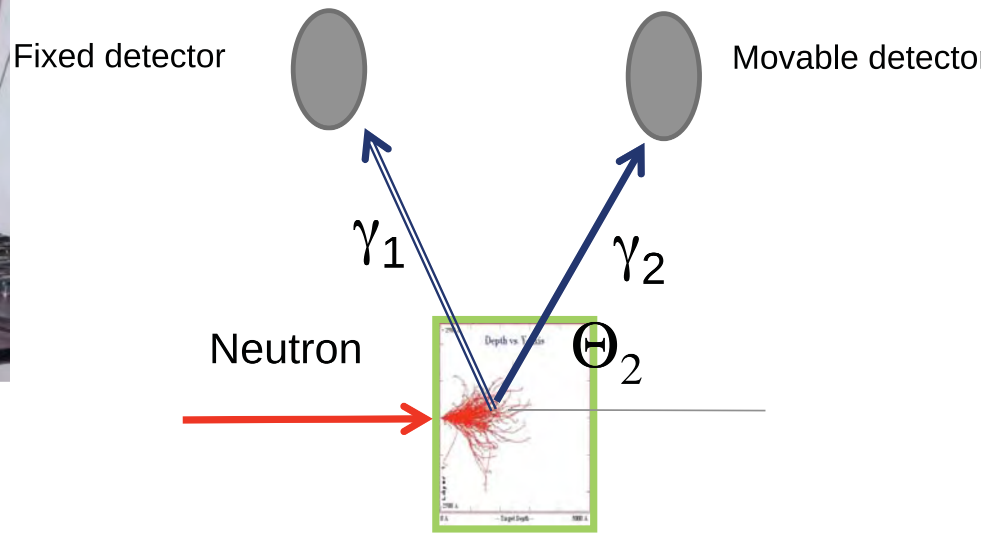
- "The Chlorine Isotope Composition of the Moon and Implications for an Anhydrous Mantle ", Z.D. Sharp, et al., et.al., Science **329**, 1050-1053 (2010)
- "Handbook of Modern Ion Beam Materials Analysis", ed. Yongqiang Wang and Michael Nastasi (December 2009)
- "Ion Implantation and Synthesis of Materials", Michael Nastasi and James W. Mayer (Springer-Verlag, Berlin, 2006)

LANSCE supports a variety of facilities for materials in radiation environments

I(rradiation) of C(hips) and E(lectronics) aka ICE House

Neutron-induced reactions on Si → p, α → ion tracks → single event effects (bit flip, latch-up)

Perturbed Angular Correlation experiment measures extent of damage cascade and local symmetries

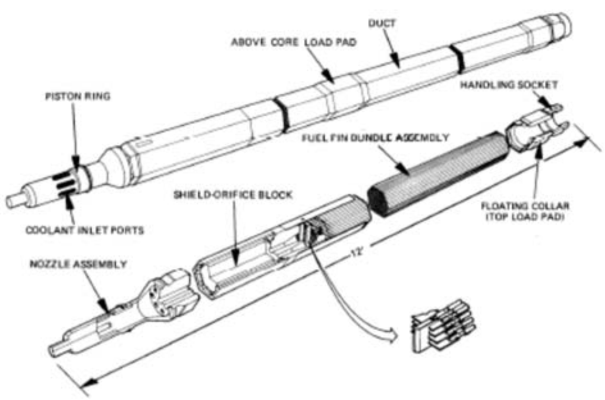


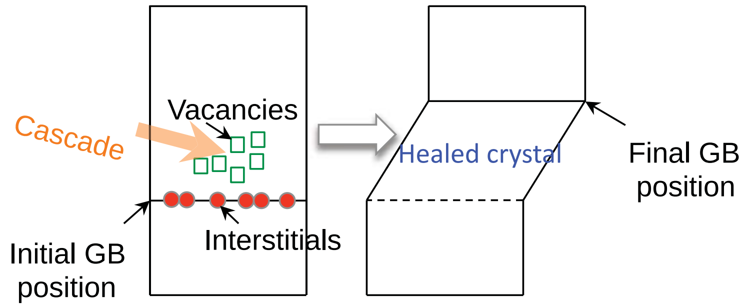
Cascade in sample

Key science questions emerged from the deep dive on materials in radiation extremes

- How do we co-design materials for nuclear energy applications?
  - Fuels/wastes to last millenia
  - Radiation tolerant materials
- What are the effects of irradiation on thermophysical properties?
- **What are the relevant time and length scales for radiation effects?**
- How do interfaces and microstructure control irradiation response of materials? And how can we control them?
- **What are the appropriate in-situ measurement capabilities for measuring damage evolution from irradiation cascades to microstructural defects?**
- What computational capability is required to model these processes over the range of length and time scales?
- **How do accelerated aging and non-accelerated aging experiments correlate?**
- What are the separations challenges that we need to “improve” to diminish nonproliferation concerns, improve economics and improve safety?

Fission and fusion reactors are major applications for research on materials in radiation environments



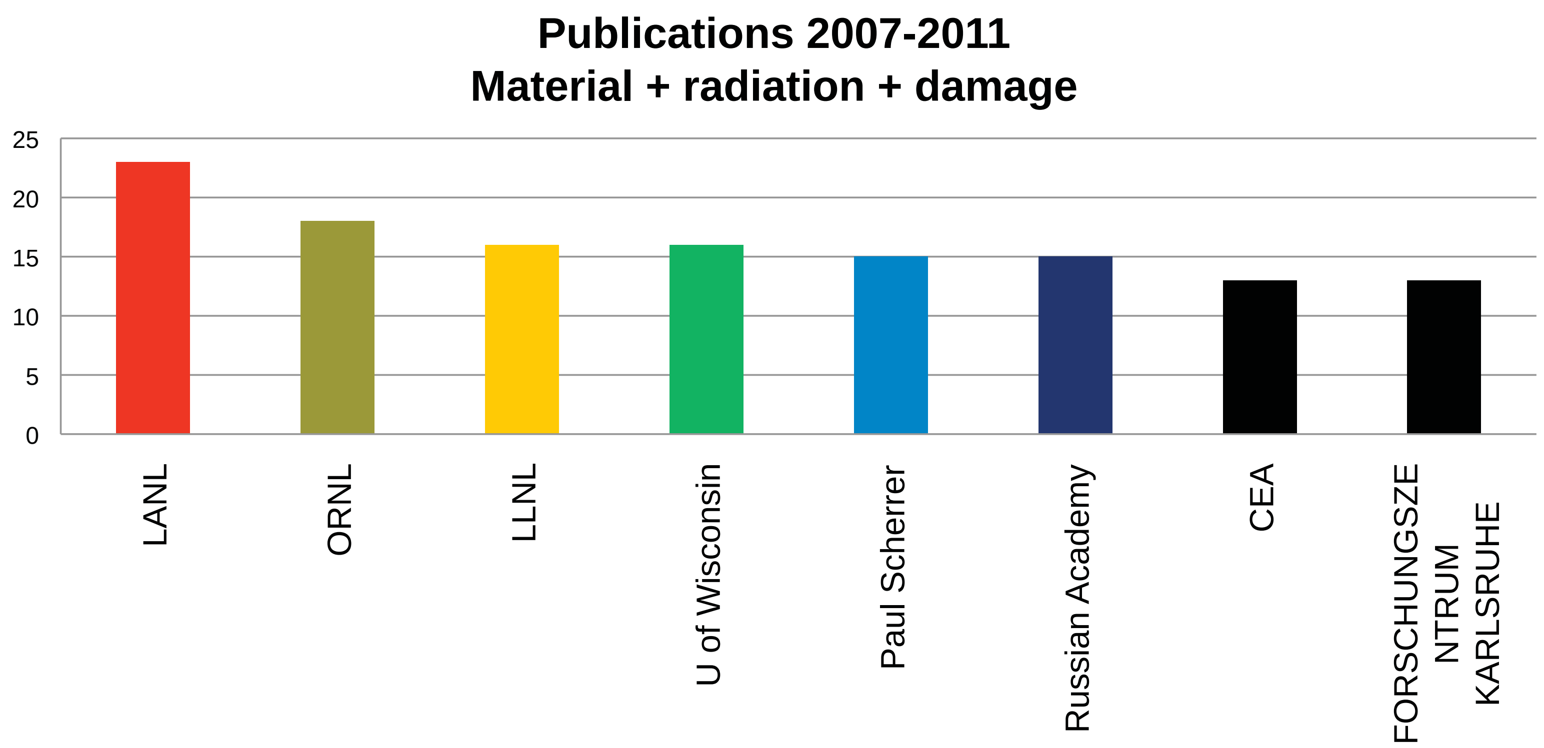


Some significant publications

- "The mechanical properties of 316L/304L stainless steels, Alloy 718 and Mod 9Cr-1Mo after irradiation in a spallation environment",S. A. Maloy, et al., Journal of Nuclear Materials **296** 119-128 (2001); Cited: 45
- "Determination of helium and hydrogen yield from measurements on pure metals and alloys irradiated by mixed high energy proton and spallation neutron spectra in LANSCE ",F. A. Garner, et al., Journal of Nuclear Materials **296** 66-82 (2001); Cited: 30
- "Mechanical properties and microstructure in low-activation martensitic steels F82H and Optimax after 800-MeV proton irradiation", Y. Dai, et al., Journal of Nuclear Materials **283** 513-517 (2000); Cited: 27
- "Extending the Time Scale in Atomistic Simulation of Materials" , A.F. Voter, et al, Annu. Rev. Mater. Res. **32** 321 (2002); Cited 232

- "Models of liquid metal corrosion", Zhang J, Hosemann P, Maloy S, Journal of Nuclear Materials **404**, 82-96 (2010)
- "Micro-structural characterization of laboratory heats of the Ferric/Martensitic steels HT-9 and T91", P. Hosemann, et al., Journal of Nuclear Materials **403**, 7-14 (2010)
- "Liquid metal embrittlement of silicon enriched steel for nuclear applications", J. Van den Bosch, et al., Journal of Nuclear Materials 398, 116-121 (2010)

With some publication metrics, LANL comes out on top of its competitors



Materials in radiation environments theme area addressed in detail in 2 presentations and 9 posters

<u>Presentations</u>

Michael Nastasi (MPA-CINT) – Center for Materials at Irradiation and Mechanical Extremes (CMIME)

Yongqiang Wang (MST-8) – Research Activities at the Ion Beam Materials Laboratory (IBML)

Materials in radiation environments theme area addressed in detail in 2 presentations and 9 posters

Posters

Eric Pitcher (LANSCE-DO) - Irradiation Environment in the Proposed Materials Test Station
John Ullmann (LANSCE-NS) – LANSCE: Nuclear Physics and Material Science

Blas Uberuaga (MST-8) - The role of grain boundaries in modifying radiation damage evolution in simple metals
Ruifeng Zhang(T-3) - Atomistic modeling and experimental studies of the shock response of Cu/Nb nanolayered composites

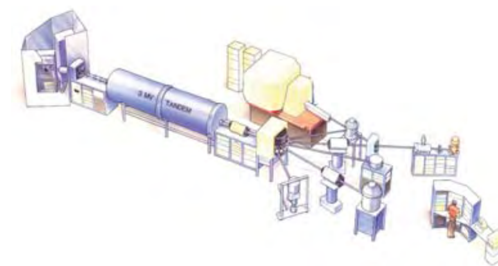
Igor Usov (MST-7) - Irradiation damage effects in ceramic oxides induced by high temperature and high fluence ion beam irradiation
Jane Burward-Hoy (ISR-1) - IBML Proton Calibration Results of the SABRS Energetic Particle Subsystem (ZEP FU1)
Jennifer Lillard (MST-6) - Effects of Radiation Environments on Long-Term Aging of Weapons Materials

Osman Anderoglu (MST-8) - Advanced Materials Development and Testing for Fuel Cycle Research & Development Program (FCRD)
Art Voter (T-1) - Influence of interstitials or vacancies introduced into grain boundary on sliding process in bcc tungsten

Research on materials in radiation environments is a vital component of LANL materials capability

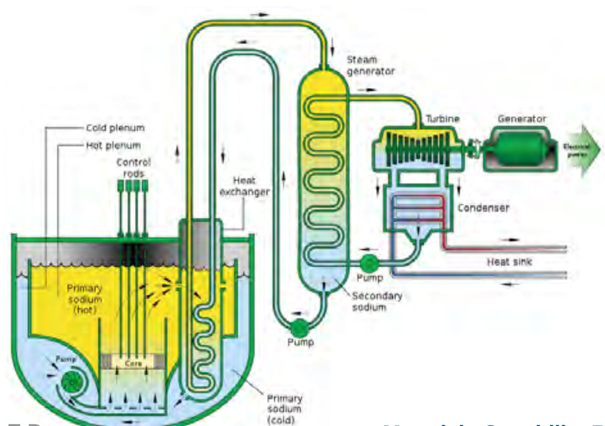
CMIME
Center for Materials at Irradiation and Mechanical Extremes

IBML



LANSCE

Fission and fusion reactors



Center for Materials at Irradiation and Mechanical Extremes

M. Nastasi (MPA-CINT)

Our new Energy Frontier Research Center (EFRC), *Center for Materials at Irradiation and Mechanical Extremes* focuses on the development of extreme environment-tolerant materials via atomic scale design of interfaces. This center recognizes that the challenge to developing materials with radically extended performance limits at irradiation and mechanical extremes will require designing and perfecting atom- and energy-efficient synthesis of revolutionary new materials that maintain their desired properties while being driven very far from equilibrium. A primary aspect associated with this challenge is to develop a fundamental understanding of how atomic structure and energetics of interfaces contribute to defect and damage evolution in materials. Recent experimental and modeling work performed in our center and presented here has shown that interface structure influences defect evolution in irradiated and mechanically damaged materials.

Center for Materials at Irradiation and Mechanical Extremes

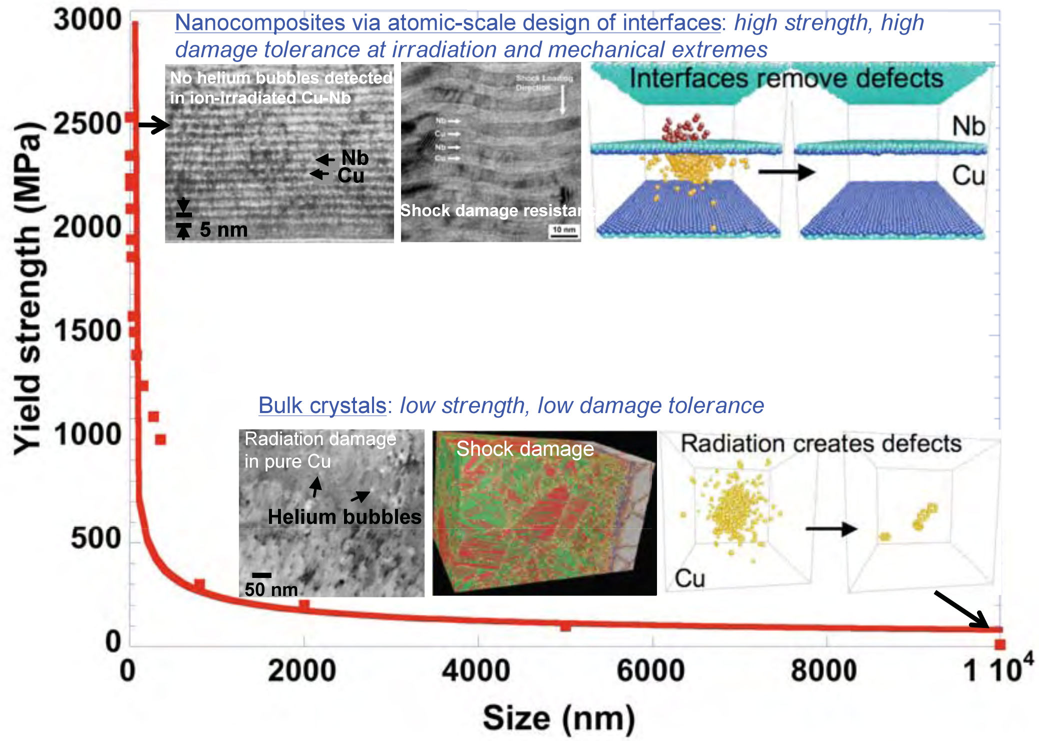
Michael Nastasi

EFRC Director

- Work supported by DOE OBES

CMIME's central idea

Atomic-scale design of interfaces can lead to tailored performance at extremes of irradiation doses and mechanical stresses



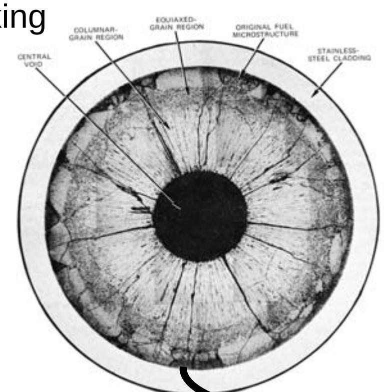
Interfaces control both the irradiation and mechanical response of the material

What is the problem?

Materials used in current nuclear reactor designs were *NOT* designed for radiation damage resistance!

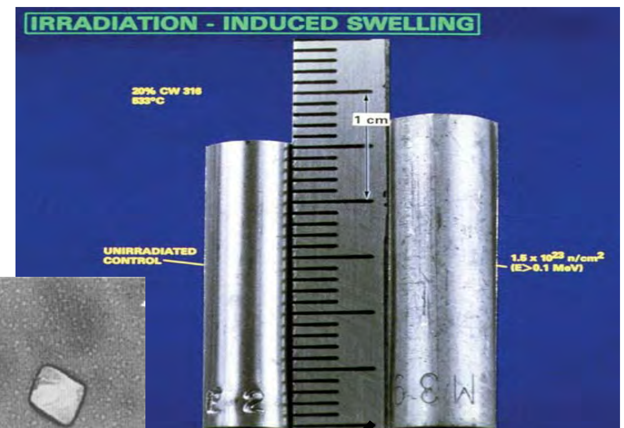
- Nuclear reactor materials chosen for strength, melting temperature, thermal conductivity, etc.
- These materials fail in unexpected ways:

Fuel pellets fail by fracture due to radiation-induced embrittlement and cracking

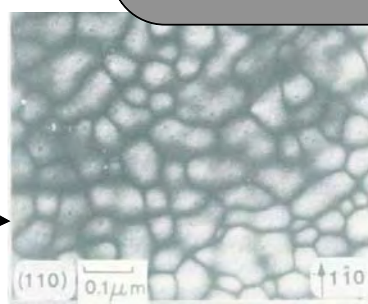


Radiation-induced point defects accelerate diffusion, form clusters, stabilize precipitates of implanted elements

Component dimensions change by >10% due to radiation-induced swelling

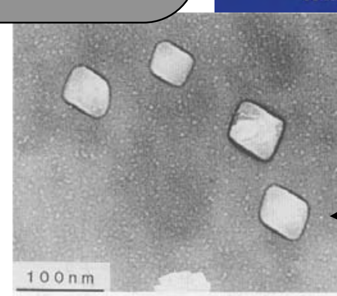


75 DPA



Segregation of solutes to grain boundaries

P. R. Okamoto and L. E. Rehn, J. Nucl.

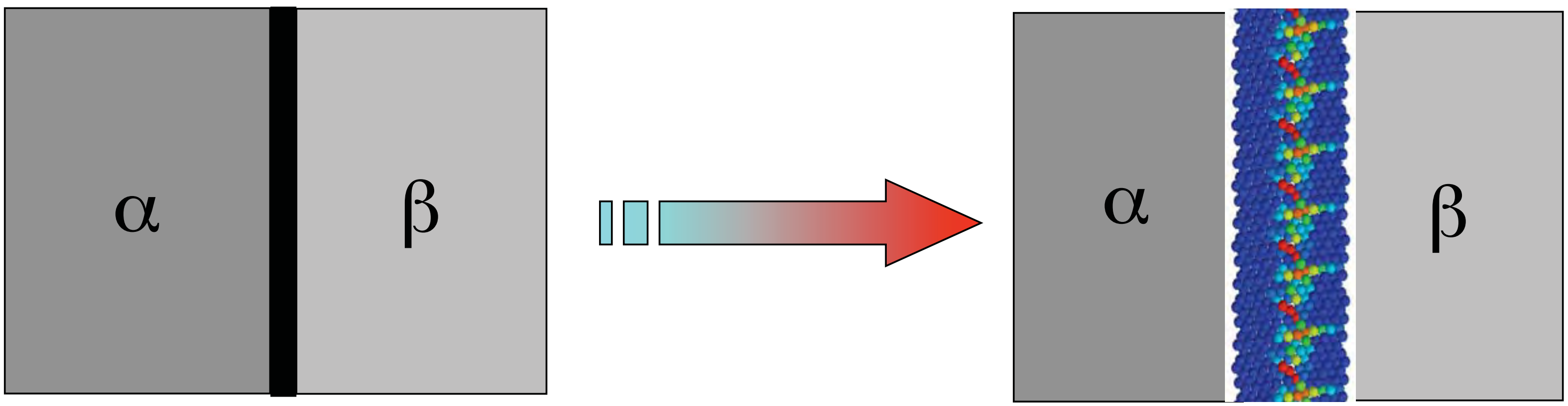


M. Victoria *et al.*, Rad. Ind. Changes

He precipitation into bubbles, formation of voids

Approach to solving this problem?

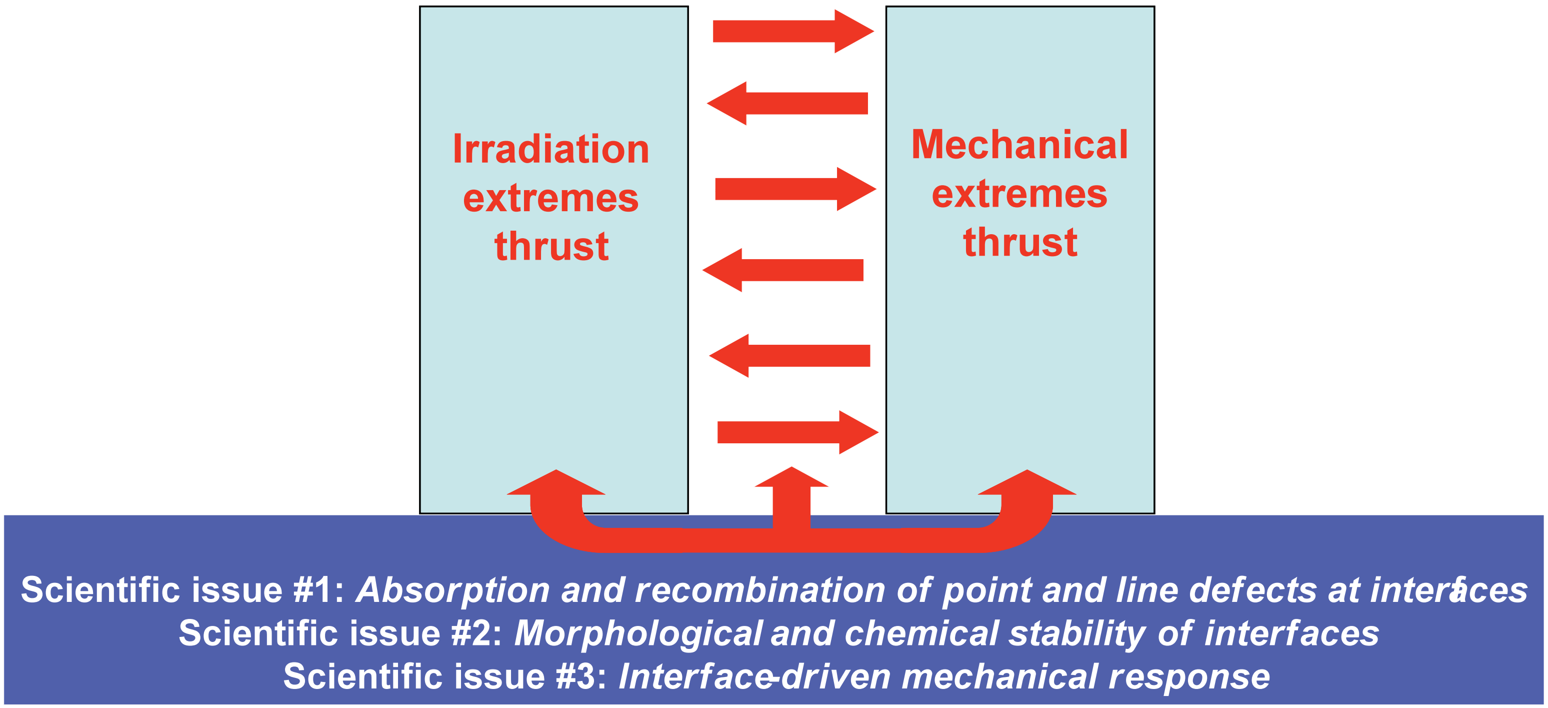
Extreme environment-tailored materials via atomic scale design of interfaces



"Materials are like people; it is the defects that make them interesting."

Key scientific issues that integrate our center

Three scientific issues (and associated hypotheses) form the unifying foundation that synergistically integrates our effort



Common Science in Both Thrust

⇒ Impact is Doubled

CMIME Org Chart

CMIME
Center for Materials at Irradiation and Mechanical Extremes

Institutions:
LANL, MIT,
UIUC, CMU

cmime.lanl.gov

Mike Nastasi, Director
Amit Misra, Co-Director

Elysha Quintana (admin)

Scientific Advisory Committee
Mike Baskes, Chair, UCSD
John Hirth, OSU (emeritus)
Bill Nix, Stanford
Bob Odette, UC-SB
Adrain Sutton, Imperial Collage
Francois Willaime, CEA/Saclay

Irradiation extremes thrust

Metals

Mike Demkowicz, MIT, lead
Kedarnath Kolluri, PD, MIT
Aurelien Vattre, PD, MIT
Liang Zhang, GRA, MIT
Alfredo Caro
Enrique Martinez, PD
Amit Misra
$*Stuart Maloy*
Osman Anderoglu (60%), PD
Mike Nastasi
Engang Fu, PD

Oxides

Blas Uberuaga, lead
Art Voter
Xian-Ming Bai (50%), PD
^Samrat Choudhary, PDF
Louis Vernon, PD (66%)
Steve Valone
*Helen Telila , GRA, UNM
$*Chris Stanek*
Aylin Karakuscu, PD
$*Quanxi Jia*
Mujin Zhuo, PD
Dzmitry Yarotski
$*Jonghan Won*
*Yun Xu, GRA, NMSU

Mechanical extremes thrust

Severe Plastic Deformation (SPD)

Irene Beyerlein, lead
^Ruifeng Zhang (50%), PDF
Nathan Mara
Tony Rollett (CMU)
Jon Ledonne, GRA, CMU
Bob Averback (UIUC)
Pascal Bellon (UIUC)
Elvan Ekiz, GRA, UIUC
Tim Lach, GRA, UIUC
$*Nhon Vo*, GRA, UIUC
$*Jian Wang*
*Thomas Nizolek, GRA, UCSB

High Strain Rates

Tim Germann, lead
^Ruifeng Zhang (50%),
Christian Brandl, PD, (50%)
Ellen Cerreta
$*Rusty Gray*
Alex Perez-Bergquist, PD, (50%)
Shengnian Luo
Weizhong Han, PD
$*Dick Hoagland*
(Lab associate)

Rob and Pat Dickerson (EML)
Yong Wang (IBML)
J. Kevin Baldwin (PVD)
Ken McClellan (solidification processing)

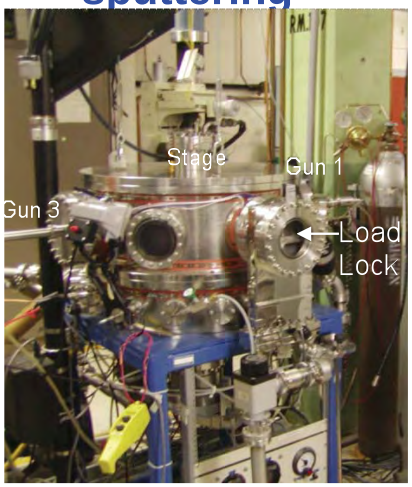
*GRA supported through *LANL Institutes*
^ LANL Director's Postdoctoral Fellowship
$collaborator, < 10% FTE on CMIME

22 staff ≈ 5 FTE
4 faculty
14 postdocs (≈ 11.5 FTE)
7 students (7 FTE); 3 supported by *LANL Institutes*

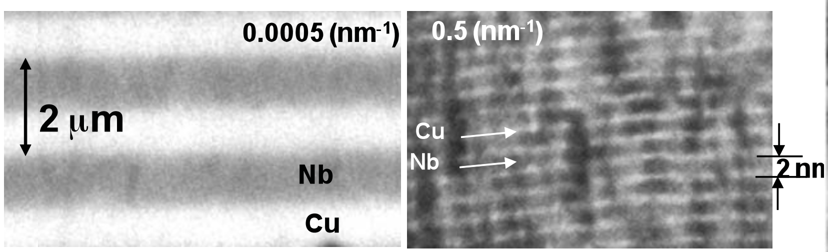
Dec 14th, 2010

Underpinning science
interfaces reduce radiation damage
by acting as sinks for radiation-induced point defects

Cu-Nb multilayer nanocomposites synthesized by magnetron sputtering



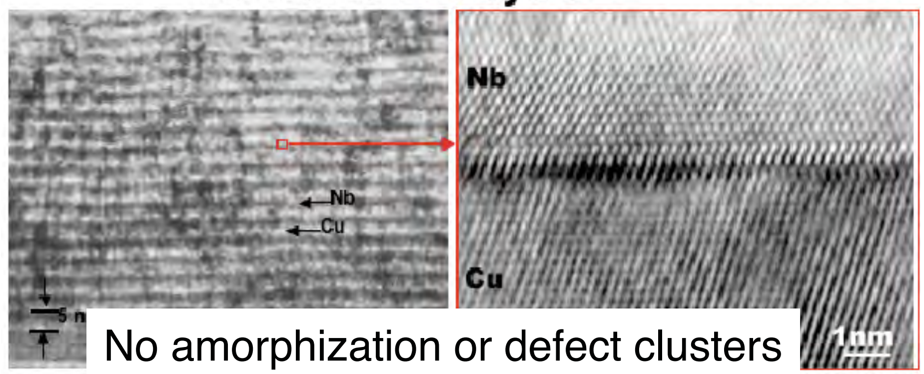
Magnetron sputtering chamber located at CINT



Overall thickness:100 nm to 100 μm

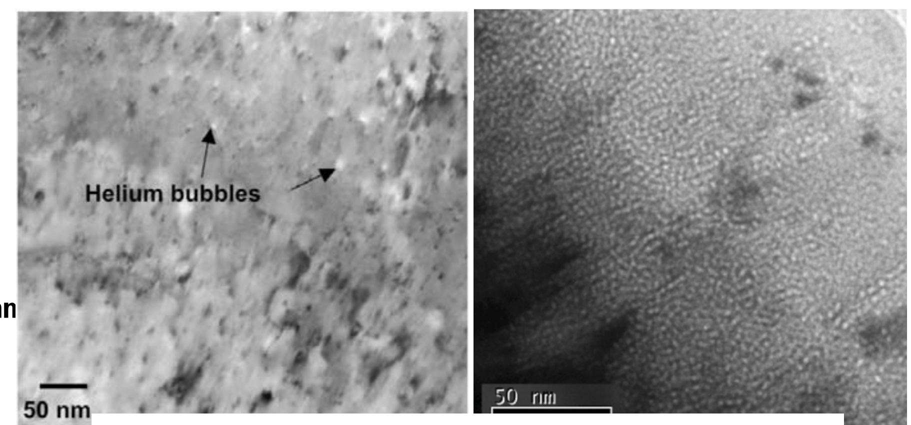
10^{17}/cm^2 150keV He implantation (~7 DPA)

Cu-Nb 2.5 nm layers



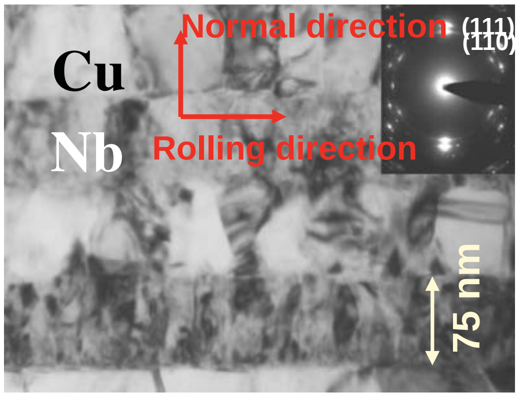
No amorphization or defect clusters

Bulk fcc Cu and bcc Nb

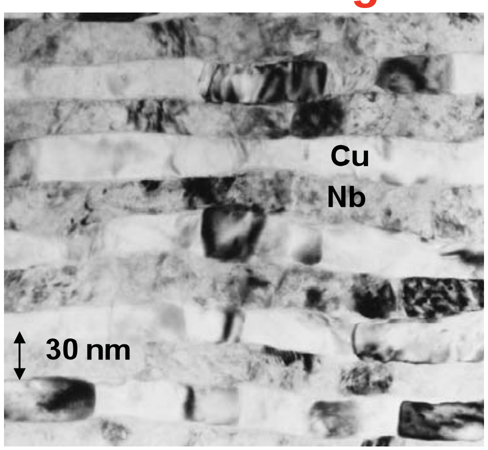


Numerous defect clusters are formed

Underpinning science
interfaces reduce mechanical damage
by acting as sinks for line defects

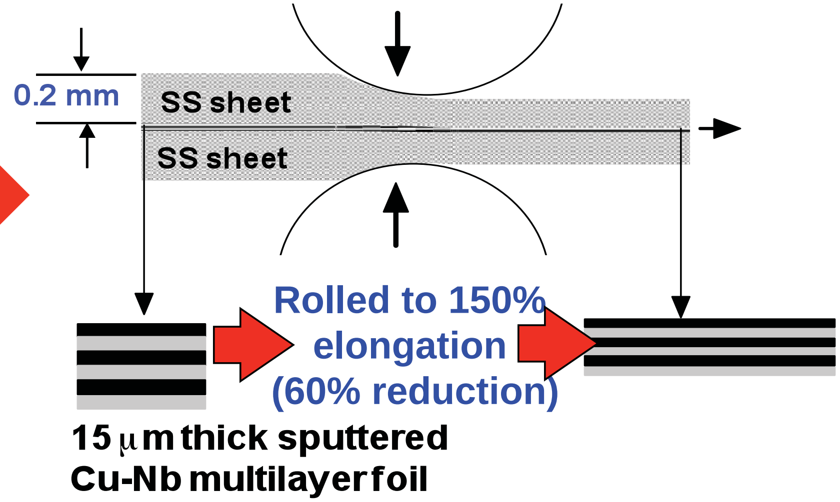


After rolling

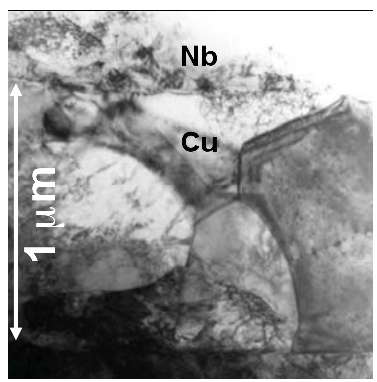


Homogeneous Deformation

Initial layer thicknesses ranged from 4 micrometers to 4 nm

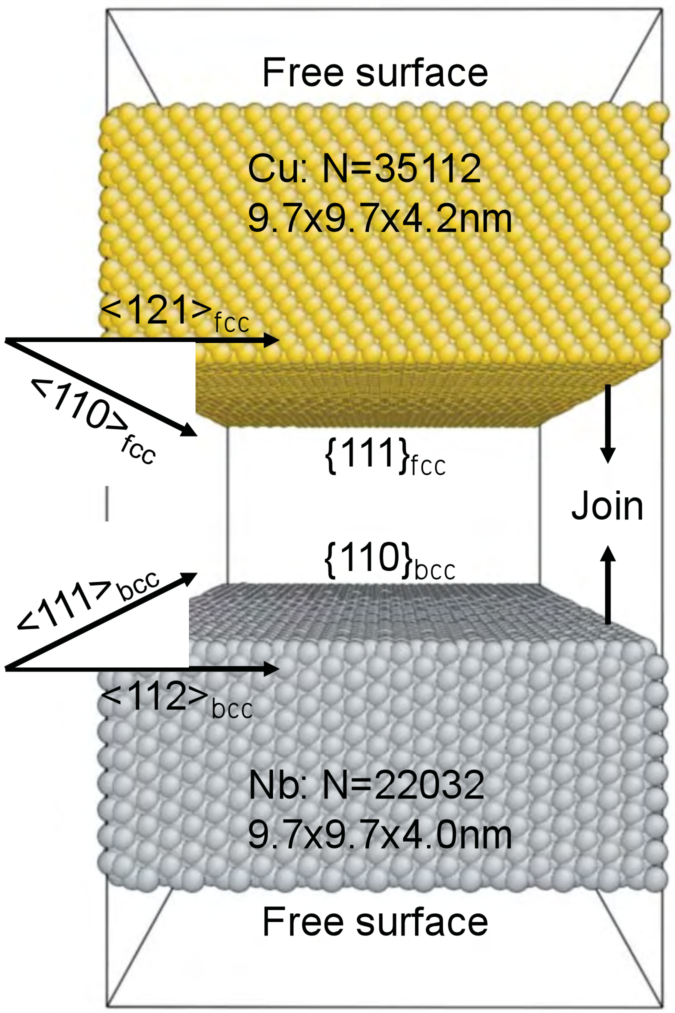


2 μm initial thickness, rolled to ≈ 50% reduction



Inhomogeneous Deformation

Basic understanding: atomistic simulation



- Join fcc-Cu and bcc-Nb in the experimentally observed Kurdjumov-Sachs (KS) orientation relation (OR)
- KS Cu-Nb interface is incommensurate
- Dimensions chosen to minimize strains needed to impose periodic boundary conditions in interface plane
- Interatomic forces modeled using an embedded atom (EAM) potential:

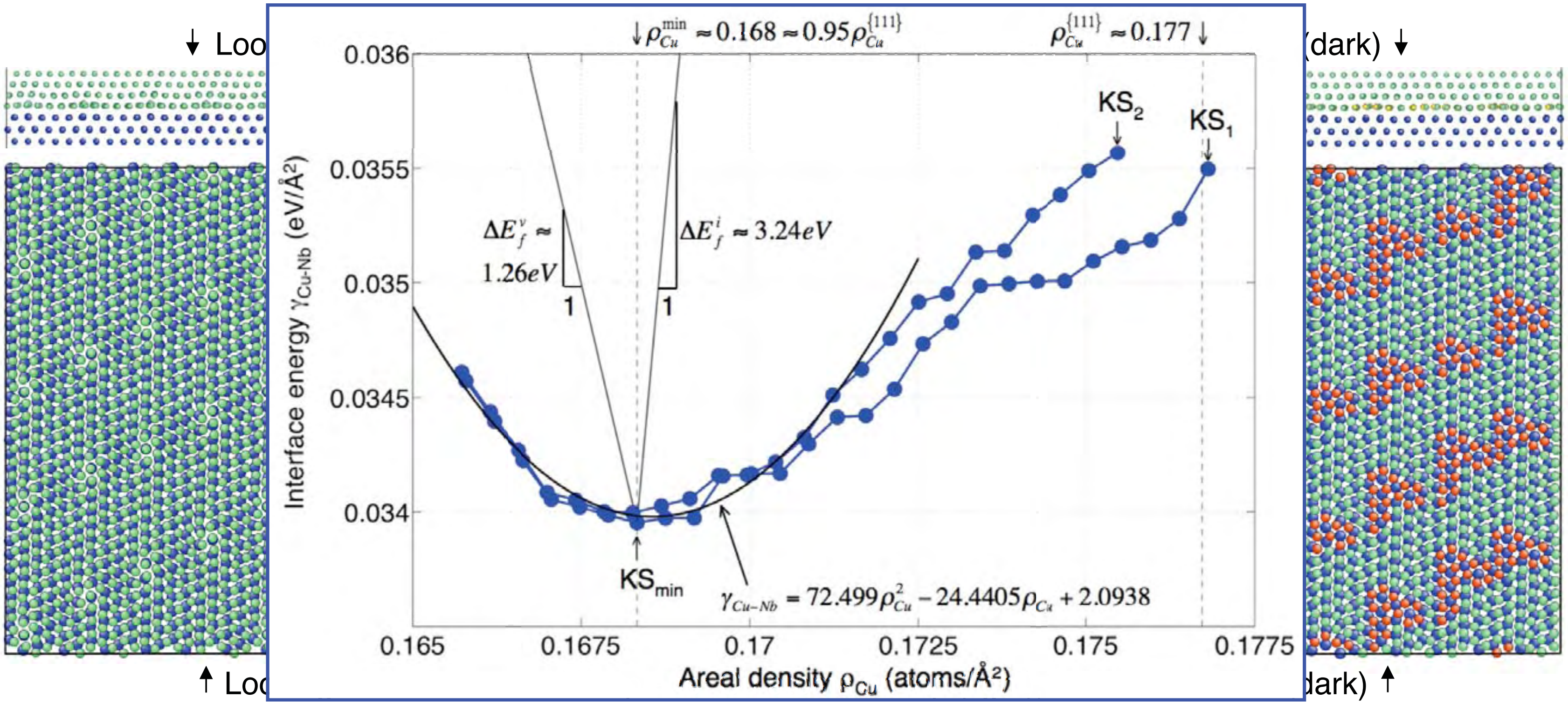
$$V_{Cu-Nb} = V_{Cu}\left[s_{Cu}, g_{Cu}\right] + V_{Nb}\left[1, g_{Nb}\right] + \sum_{i_{Cu}, i_{Nb}} \phi_{Cu-Nb}\left(\vec{r}_{i_{Cu} i_{Nb}}\right)$$

  – Cu: A. F. Voter
  – Nb: R. A. Johnson and D. J. Oh
  – Inter-element interaction reproduces:

Multiple interface atomic configurations

each in the Kurdjumov-Sachs orientation relation with nearly degenerate energies

KS_1	KS_2	KS_{min}
• Created by simply joining Cu and Nb in the KS OR • Quasiperiodic pattern of low coordination sites	• Interfacial Cu atom layer homogeneously strained with respect to Cu (111) • No low coordination sites	• Contains a 5% atomic vacancy concentration • Lowest energy configuration at T=0K

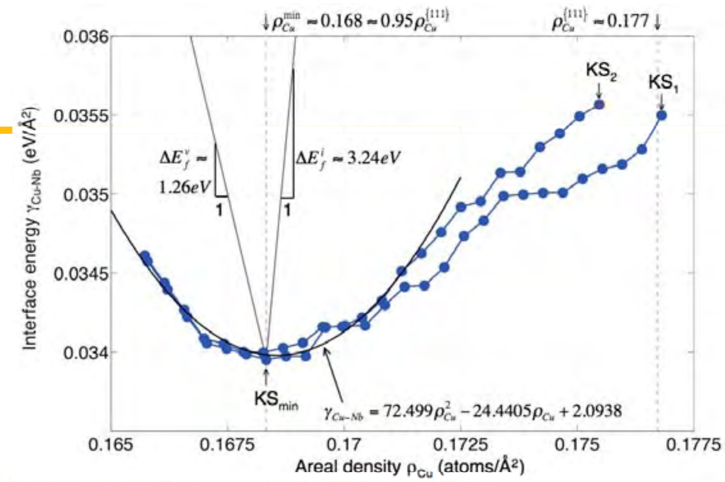


Consequences of interfaces with nearly degenerate energies

- Low defect formation energy

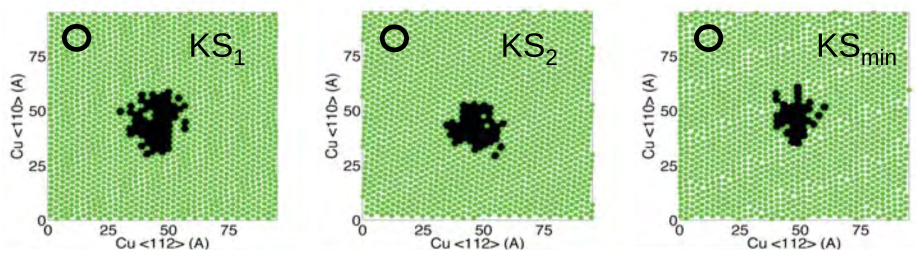
Effective formation energy for defects in KS_{min}:

$\Delta E_{eff} \approx 0.12$ **eV/defect**



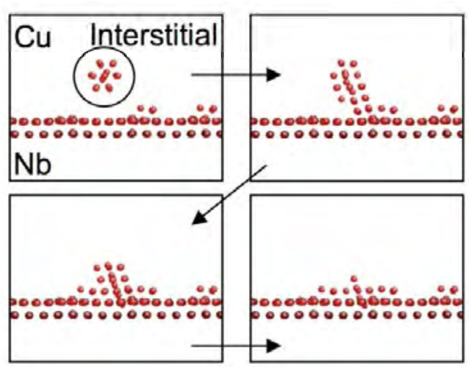
- Large defect core sizes

Effective interfacial defect recombination radius ≥ 0.75nm

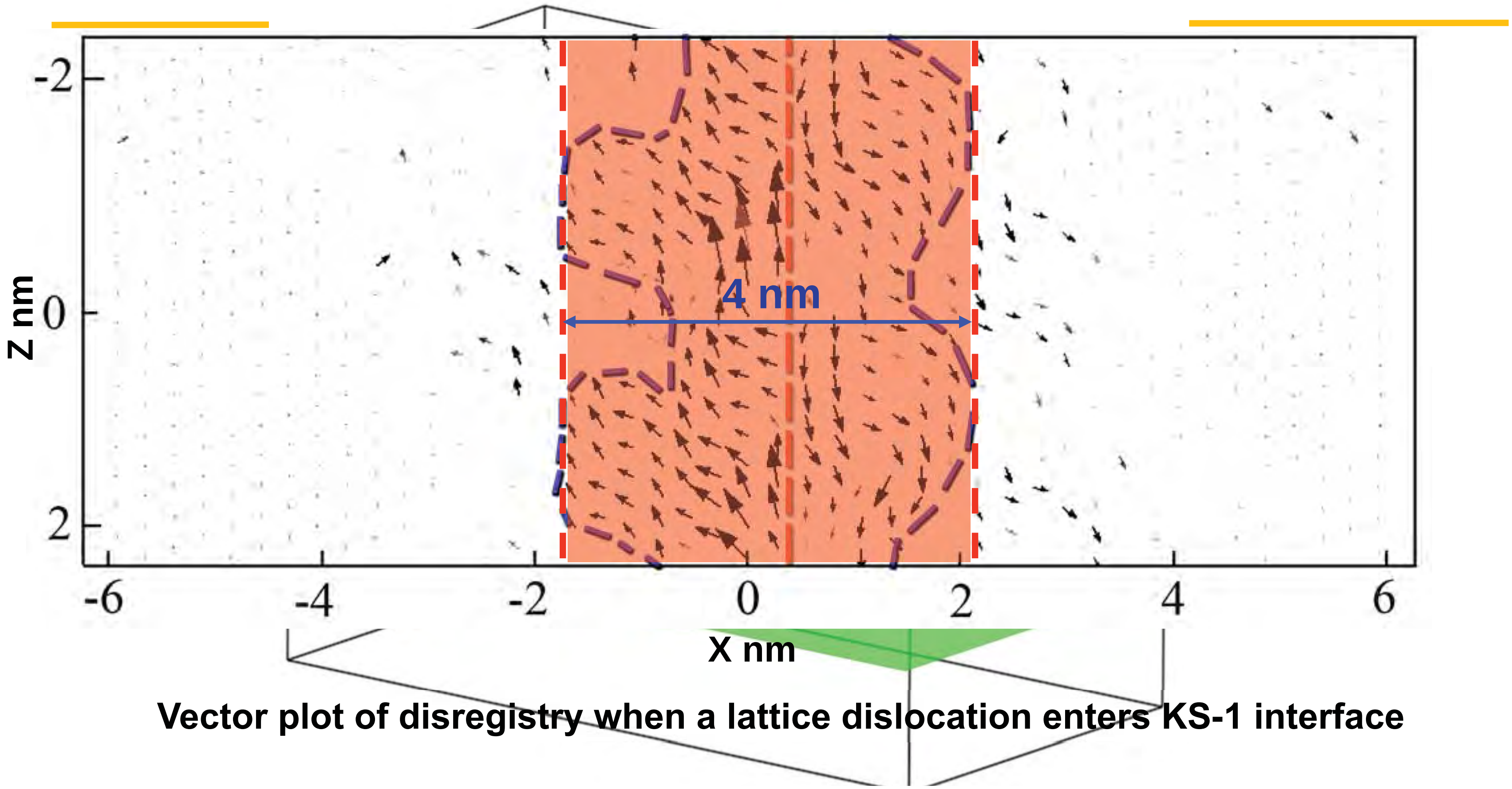


- Athermal migration of defects to interfaces

Energy barriers for point defect migration to Cu-Nb interfaces vanish at a critical distance



“Weak” interfaces are readily sheared under the stress field of a glide dislocation => attraction, absorption and core spreading in the interface plane



Vector plot of disregistry when a lattice dislocation enters KS-1 interface

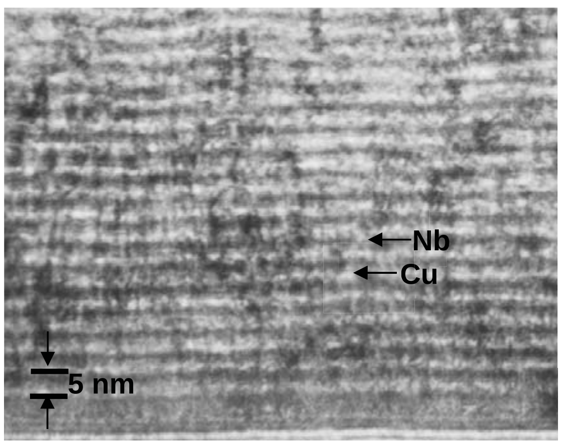
A Cu/Nb KS_1 model in which one of the Shockley dislocations has entered the interface.

J. Wang et al, Acta Mater 56:3109; 56:5685 2008

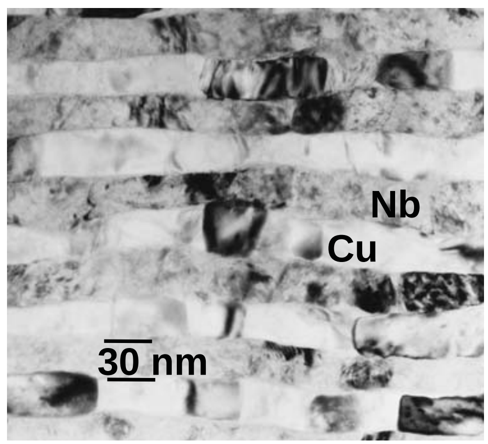
Interfaces provide damage resistance

Nanolayered metallic composites (e.g., Cu-Nb) resist damage accumulation at irradiation as well as mechanical extremes. Thus, the same interfaces can be used in a synergistic way by both thrusts.

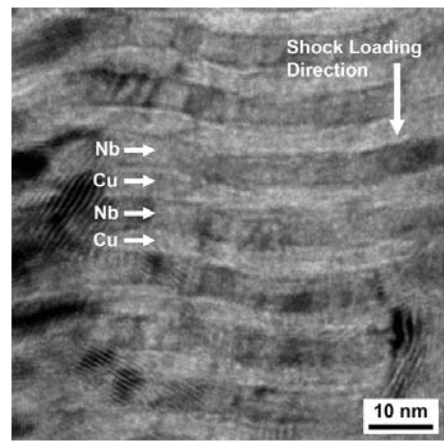
Ion irradiated



Rolled to large plastic strains



Laser shocked



Synergy through common model systems and synthesis methods

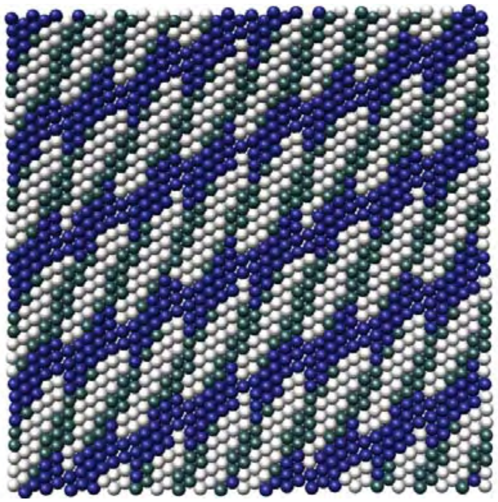
Synergy through hypotheses-driven investigations based on quantitative figures-of-merit

Interface Sink Strength: A Figure-of-Merit Example

Interfaces as Super Sinks for Defects

Generalization for any multiphase material

Modeling of simple system



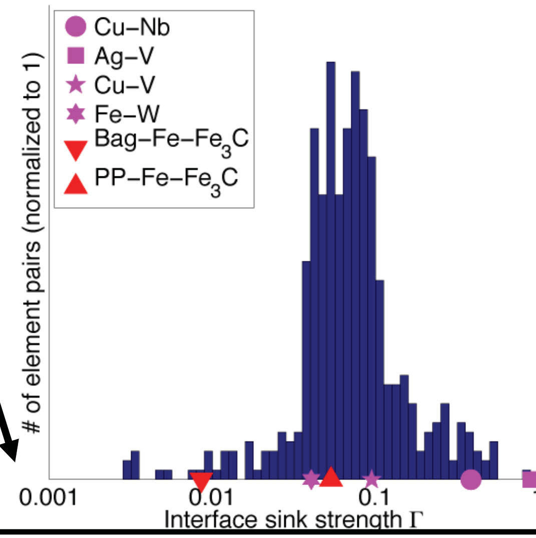
Quantification of interface sink strength

Interface sink strength (the higher the better)

$$\Gamma = \rho/\lambda$$

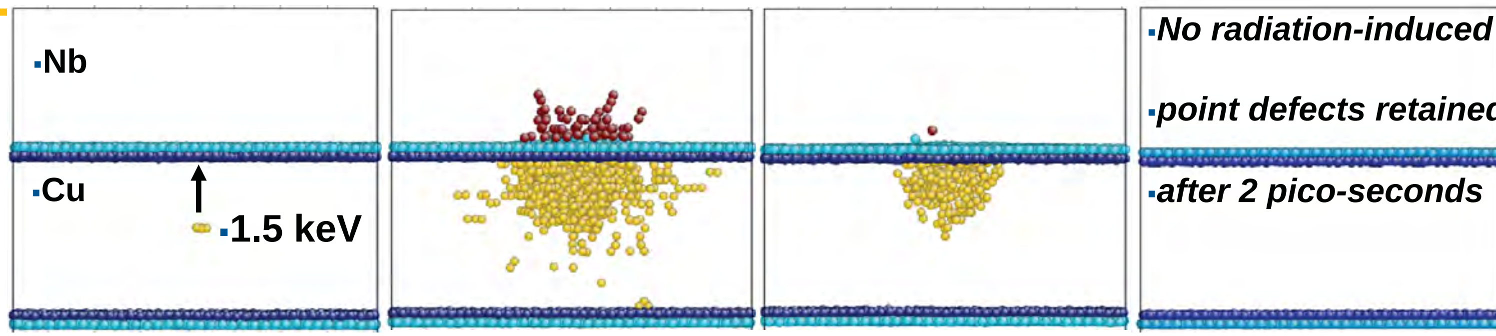
Can be used to predict what composites contain supersink interfaces

Design of composites for radiation tolerance



Evolution of the figure-of-merit cross-cuts the entire EFRC

Effects of interface structure on He solubility



- *Atomistic modeling showed that lattice misfit creates interfaces that are efficient sinks for radiation-induced point defects and catalytic sites for fast annihilation of vacancy-interstitial pairs*

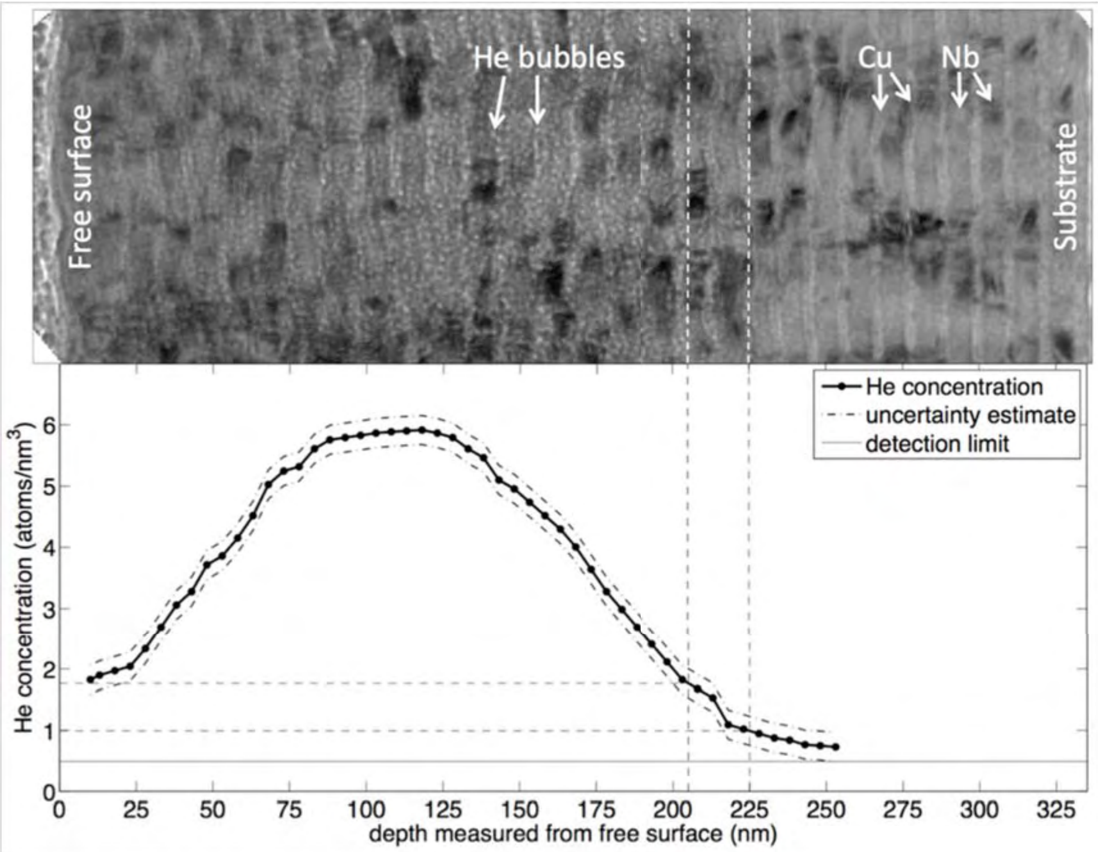
- **Defect formation energies are significantly lower in the interface than elsewhere in the crystal (0.1-0.3eV in the interface as opposed to 1-3eV in bulk).**

- **The defect mobility at interfaces is higher by at least 5 orders of magnitude than in the bulk**

- **The range of interaction with other point defects, the so-called core size of trapped defects, is ~ 6 times larger at interfaces than bulk.**

Does the interface structure influence He solubility?

Critical concentration to observe He bubbles at fcc-bcc interfaces



Systems studied

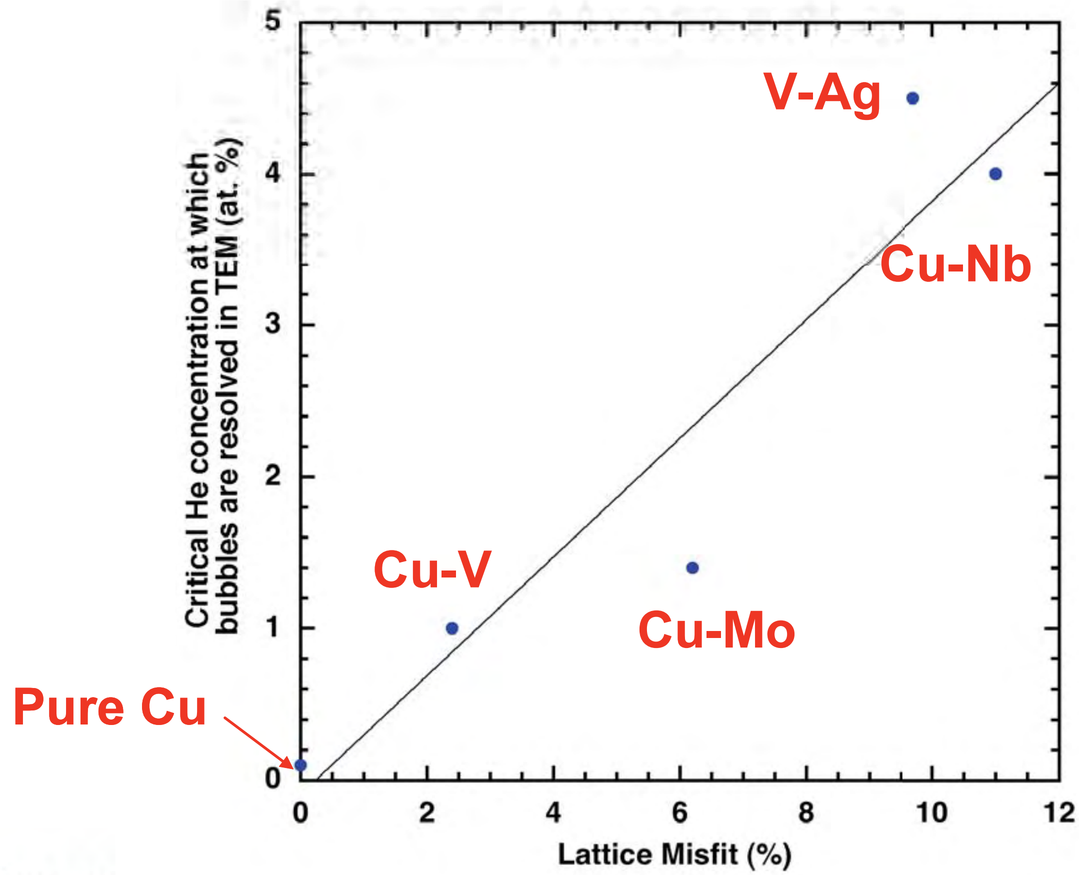
V-Ag (ε = 9.7%), Cu-Nb (ε = 11%), Cu-Mo (ε = 6.2%), Cu-V (ε = 2.4%).
(all systems are immiscible)

Σ3{111} coherent twin boundary in Cu (ε = 0%).

M. J. Demkowicz *et al.*, Appl. Phys. Lett. **97**, 161903 (2010)

(Work supported in part by LDRD DR)

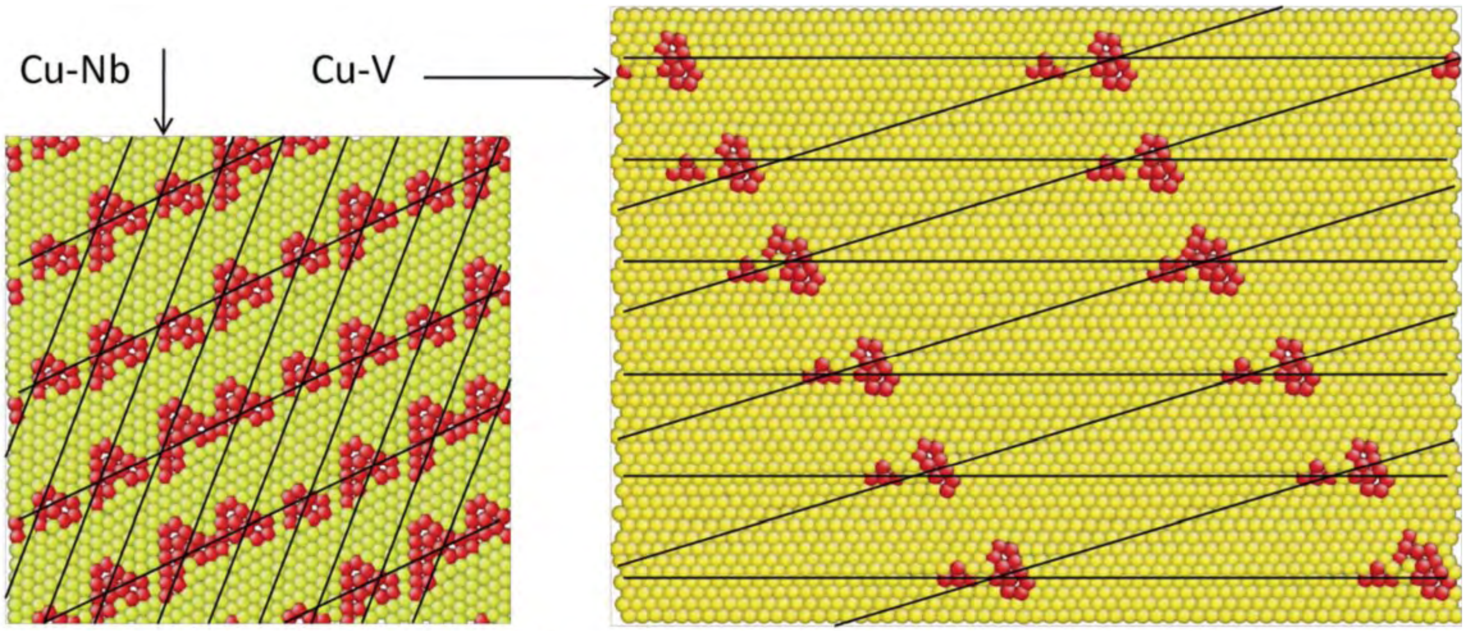
He solubility scales with lattice misfit



(Work supported in part by LDRD DR)

Interface structure is important: Constitutional vacancies at Cu-Nb and Cu-V interfaces

- Both Cu-Nb and Cu-V interfaces contain excess free volume due to ~2.5 constitutional vacancies per misfit dislocation intersection
- The areal density of misfit dislocation intersections is higher in Cu-Nb than in Cu-V, so the overall vacancy concentration is ~5% at. in Cu-Nb and ~1%at. in Cu-V
- ***This may explain why Cu-Nb has ~4.5 times higher c_{He} than Cu-V***



Red atoms indicate high free volume areas containing constitutional vacancies

CMIME is off to a flying start

- We are a CENTER
- Synergistically integrated via scientific issues, hypotheses, synthesis of common model systems…
- We are fully staffed
- New resources (Titan, Helios, cluster, PVD system)
- Led and successfully worked with other EFRCs for 1st Summer School
- Building community of early-career scientists

Current research activities at the Ion Beam Materials Laboratory

Y. Wang (MST-8)

This talk attempts to give a brief overview on current ion beam materials research activities centered around the Ion Beam Materials Laboratory (IBML) of the Materials Science and Technology Division at Los Alamos. The IBML was established in 1986 with the installation of two accelerators: a new NEC 3-MV Pelletron tandem ion accelerator and a used Varian DF-3000 production ion implanter. In 2008, a new Danfysik high current research implanter was installed with the support of the DOE Office of Basic Energy Sciences.

After a brief introduction of IBML's history, mission, and current capabilities, I will give a few highlights on current ion beam materials research activities, ranging from application of channeling nuclear reaction analysis for helping understand metal hydride behavior, to ion irradiation induced lattice swelling in titanate pyrochlore structure, to in situ quantitative nano-compression testing of ion irradiated metals, and to mimicking solar wind bombardment on lunar rocks with energetic proton particles. Finally, I will give an outlook of potential future research activities where unique IBML capabilities such as dual-beam irradiation and irradiation and corrosion experiment (ICE) capabilities could play important roles in both fundamental radiation damage science and ion beam materials research with nuclear energy applications.

Current Research Activities at Ion Beam Materials Laboratory (IBML)

Yongqiang Wang

Ion Beam Materials Laboratory
Materials Science in Radiation and Dynamics Extremes Group
Materials Science and Technology Division

Los Alamos National Laboratory

IBML sponsors and users

Mike Nastasi, LANL
Kurt Sickafus, LANL
Joe Tesmer, LANL
Amit Misra, LANL
Stuart Maloy, LANL
Carol Haertling, LANL
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Haiyan Wang, TAMU
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Wei Pan, Sandia
Mike Demkowicz, MIT
J.K. Lee, Univ. of Pittsburg
Yuhong Li, Lanzhou Univ.
Jian Zhang, Xiamen Univ.
Feng Ren, Wuhan Univ.

Outline

IBML: A dedicated multiuser research facility

Current research activities: highlights

- ✓ Understanding physics of palladium hydride behavior
- ✓ Pre-amorphization swelling in ion irradiated titanate pyrochlore oxides
- ✓ In situ nano-compression testing on ion irradiated Cu under TEM
- ✓ Mimic solar wind impact on Moon rocks: Effect on $^{37}Cl/^{35}Cl$ ratios

Future research plans

History and mission

Ion Beam Facility (IBF): (Decommissioned in early 1990s)

- ✓ Two Accelerators: Van De Graaff (Vertical) + FN Tandem
- ✓ Nuclear Physics Research Facility (protons, deuterons, tritons, alphas, etc.)
- ✓ Largely funded by weapons science programs

Ion Beam Materials Laboratory (IBML): (Established in 1986)

- ✓ 3 MV NEC Tandem Accelerator, 200 kV Varian Implanter
- ✓ One of the earliest accelerator labs dedicated for materials research

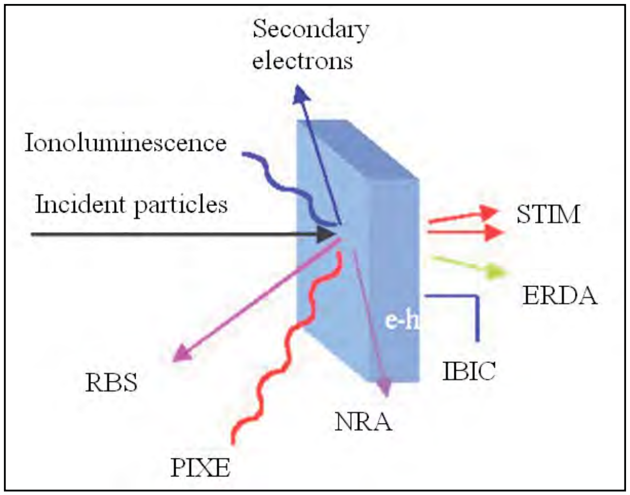
Mission:

- ✓ **Support and conduct** cutting-edge research in ion-solid interactions, ion beam analysis, ion implantation, and radiation damage effects;
- ✓ Provide centrally accessible **ion beam instrumentation** for materials modification and characterization for LANL researchers, maintained and upgraded by experts;
- ✓ Build, preserve and upgrade the **knowledge and skills** required for the optimal operation and customized research capability in the ion beam materials research;
- ✓ **Train and teach** LANL researchers, postdocs, and students to skillfully apply ion beam techniques and instrumentation to their research; and
- ✓ Provide ion beam services to DOE **CINT users** and other entities **external** to LANL.

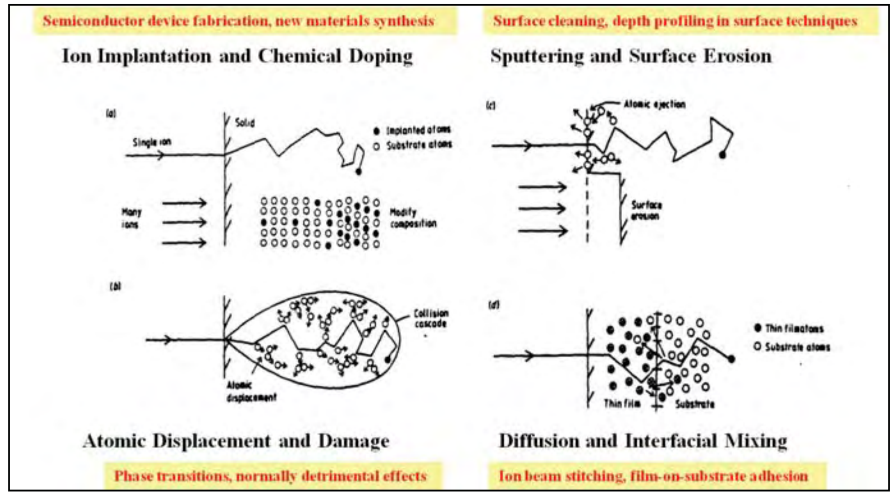
IBML contributes to materials research

Ion–solid interactions

Materials Analysis



Materials Modification and Radiation Damage

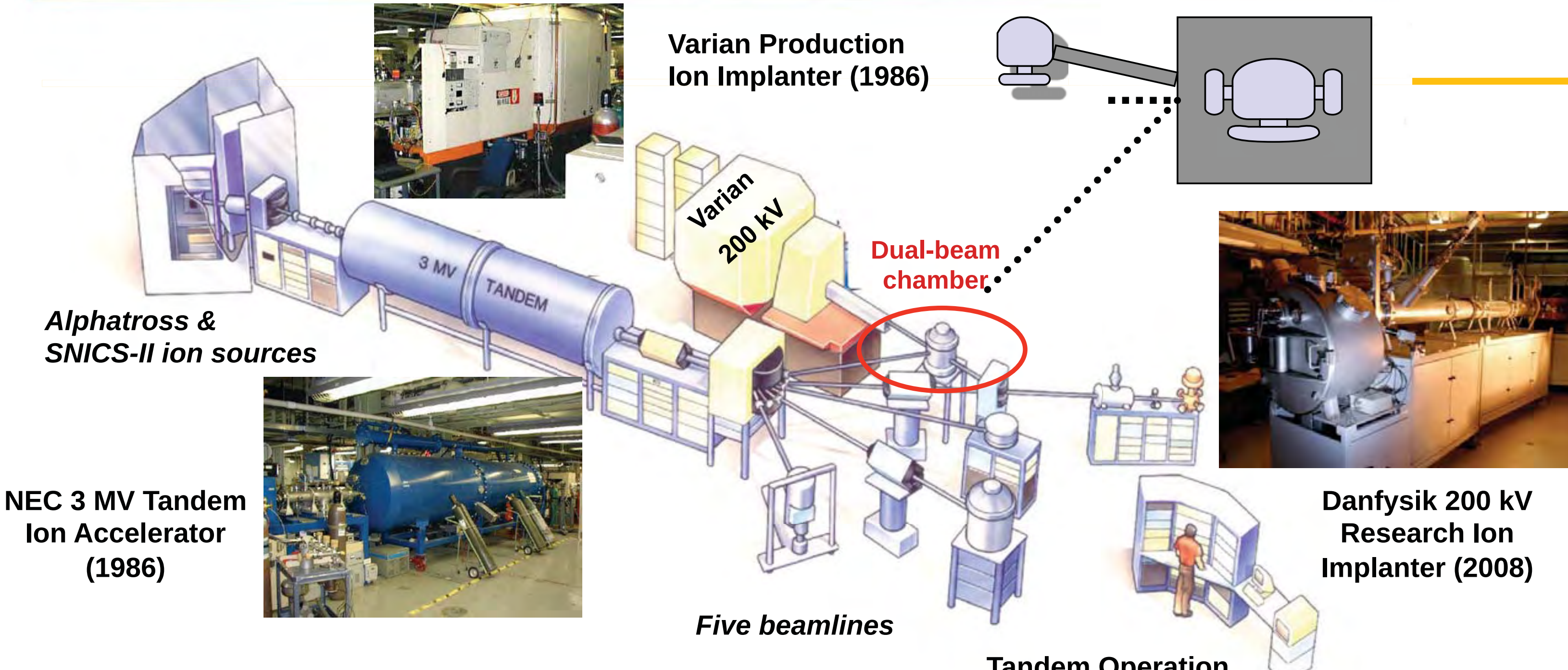


Ion beams contribute to materials research in three categories:

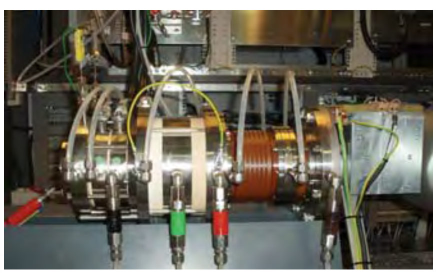
✓ Materials characterization with **ion beam analysis (IBA)** techniques
✓ Materials modification and synthesis through **ion implantation**
✓ **Radiation damage effects** in materials by ion bombardment

Ion Beam Materials Laboratory

IBML Los Alamos

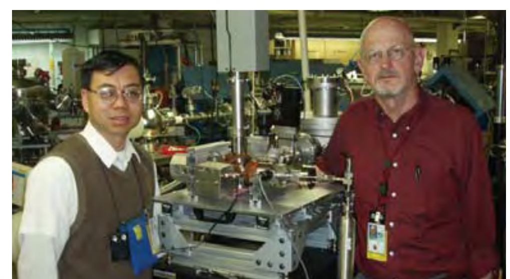


Danfysik: first of its kind in North America (BES at 2008)



Gas-Oven-Sputter Ion Source

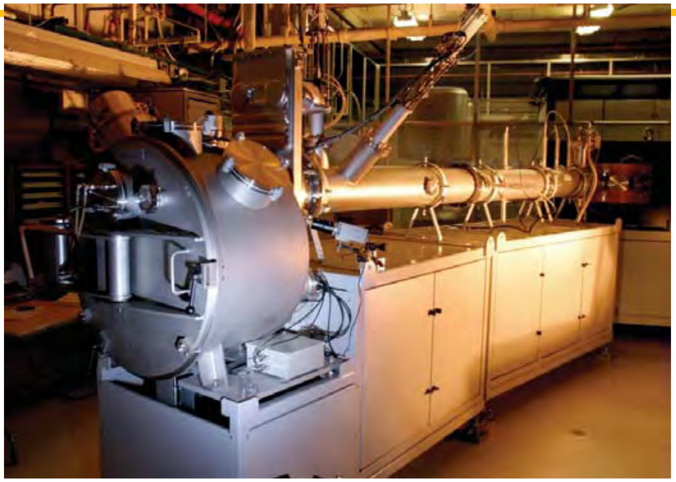
Three beamline ports magnet



Ion beam materials research capabilities at LANL

NEC 3 MV Tandem Ion Accelerator

Danfysik 200 kV High Current Ion Implanter



High Energy Ion Beam Capabilities:

- Gas & sputter ion sources and five beamlines
- Proton beam: 200 keV to 6 MeV
- Alpha beam: 200 keV to 9 MeV
- Heavy ions (C, Si, Cu, Au): 200 keV to 20 MeV
- Beam currents: from a few pA to a few μA
- Ion Beam Analysis (RBS/NRA/PIXE/channeling)
- High temperature ion irradiation (1500ºC)
- Ion irradiation and corrosion (ICE) experiment
- A tunable actinide alpha source: ~mCi to ~kCi
- Nuclear microprobe analysis and modification

High Current Ion Beam Capabilities:

- Gas-Oven-Sputter source and up to three beamlines
- Proton beam: 5 keV to 200 keV
- Helium beam: 10 keV to 400 keV
- Heavy ions (He, Ar, Fe, Xe, Au): 30 keV to 800 keV
- Beam currents: a few μA to a few mA
- Target implantation temperature: LN_2 to 500ºC
- In situ characterization: HREED
- Materials synthesis and modification
- Radiation damage science through ion irradiation
- Surface metallurgical engineering

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- ✓ Mimic solar wind impact on Moon rocks: Effect on $^{37}Cl/^{35}Cl$ ratios

Future research plans

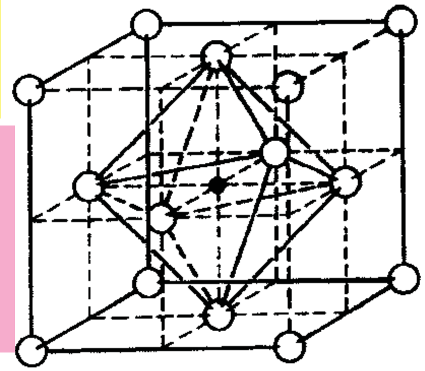
Understanding physics of palladium hydride behavior

Channeling NRA to determine ^{3}He or ^{2}D locations in Pd lattice

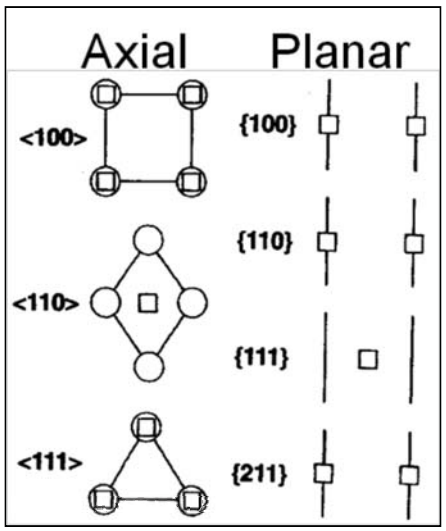
(Courtesy of D. Safarik and R. Schwarz)

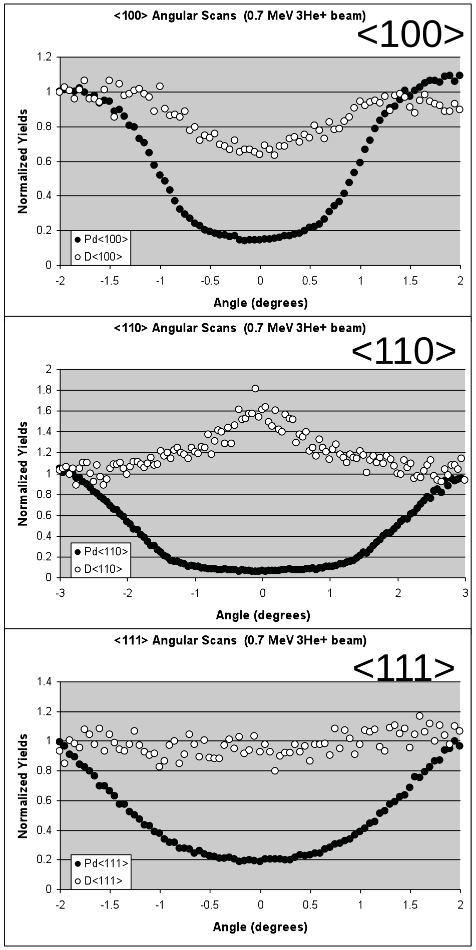
- ✓Relevant to hydrogen economy
- ✓Important to defense applications

Pd Lattice: fcc structure
$PdT_{0.8}$: ^{3}T at octahedral site
$^{3}T \rightarrow {}^{3}He + \beta^{-}$ + anti-neutrino
Does ^{3}He occupy the same ^{3}T site?



H at octahedral site





Method:

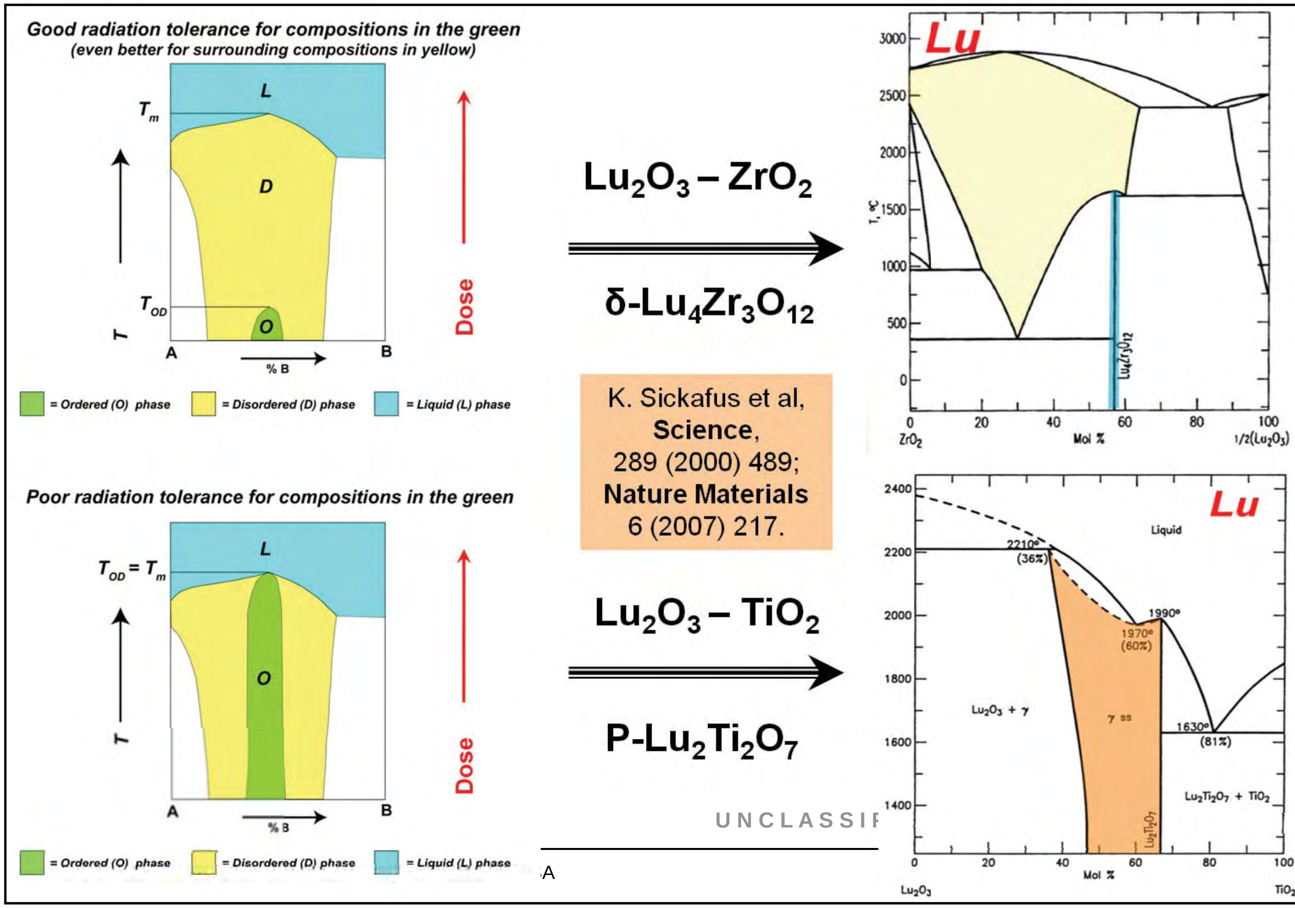
✓Using D to substitute T to form PdD: using channeling NRA $^{2}H(^{3}He,p)^{4}He$ to determine D (thus T) lattice site

✓After tritium decay, PdT to become $PdT(^{3}He)$; then isotope exchange to form $PdH(^{3}He)$: using channeling NRA $^{3}He(d,p)^{4}He$ to determine ^{3}He lattice location in Pd lattice

Pre-amorphization swelling in ion irradiated titanate pyrochlore ($Ln_2Ti_2O_7$)

(BES project, PI: K. Sickafus)

✓Potential nuclear waste form applications
✓Pyrochlore phase nanoparticles found in ODS structural materials



Excellent resistance to amorphization

Very poor resistance to amorphization

δ-$Lu_4Zr_3O_{12}$ vs. P-$Lu_2Ti_2O_7$

δ-$Lu_4Zr_3O_{12}$ has an excellent resistance to irradiation induced crystalline to amorphous (C-A) transformation since it has a strong propensity to first adopt order-disorder phase transformation via the formation of cation antisite defects in the lattice.

Questions:

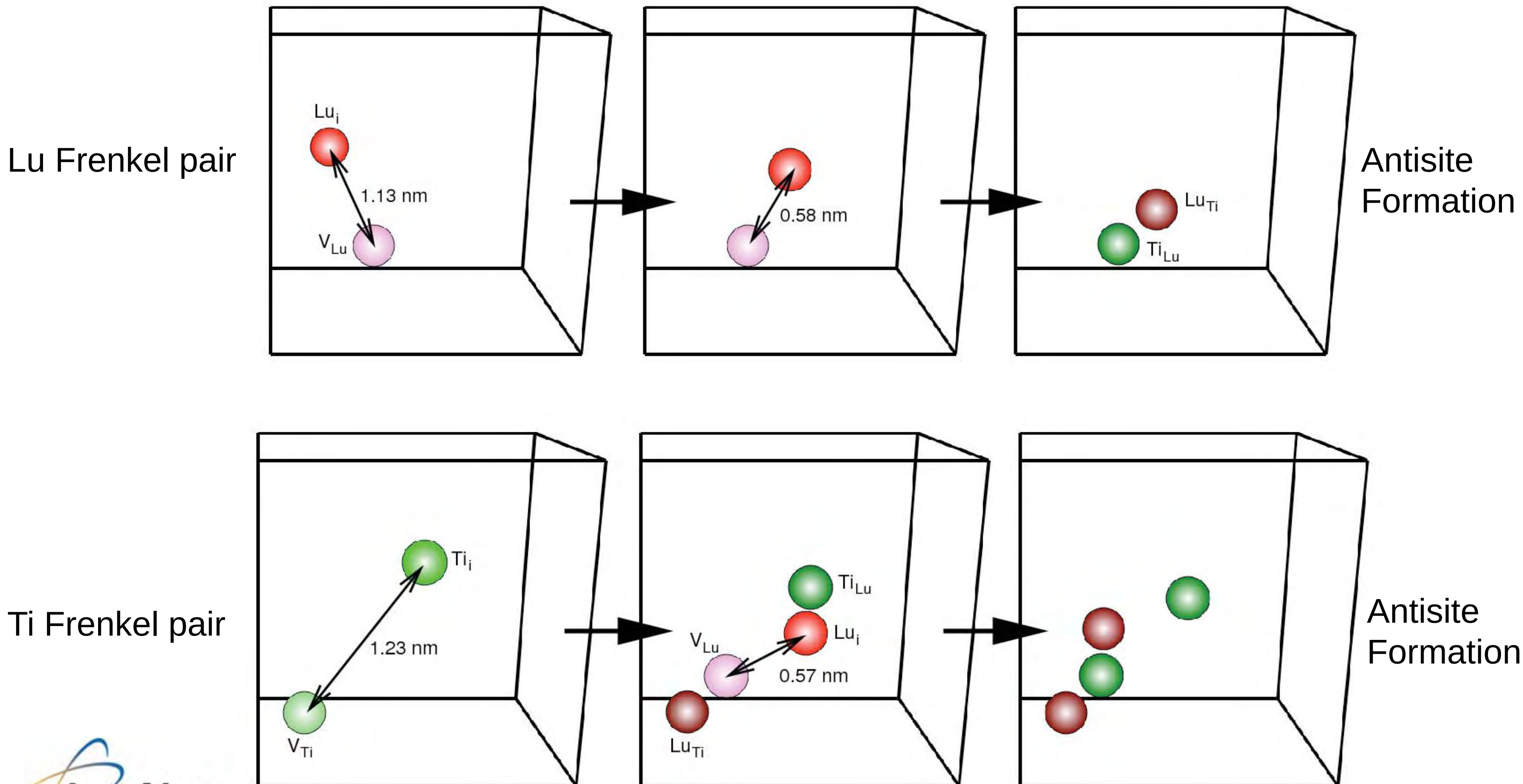
How does titanate pyrochlore (P-$Lu_2Ti_2O_7$) undergo C-A transformation?

Does it also undergo O-D phase transformation first?

If so, what type(s) of defects contribute to the O-D transformation?

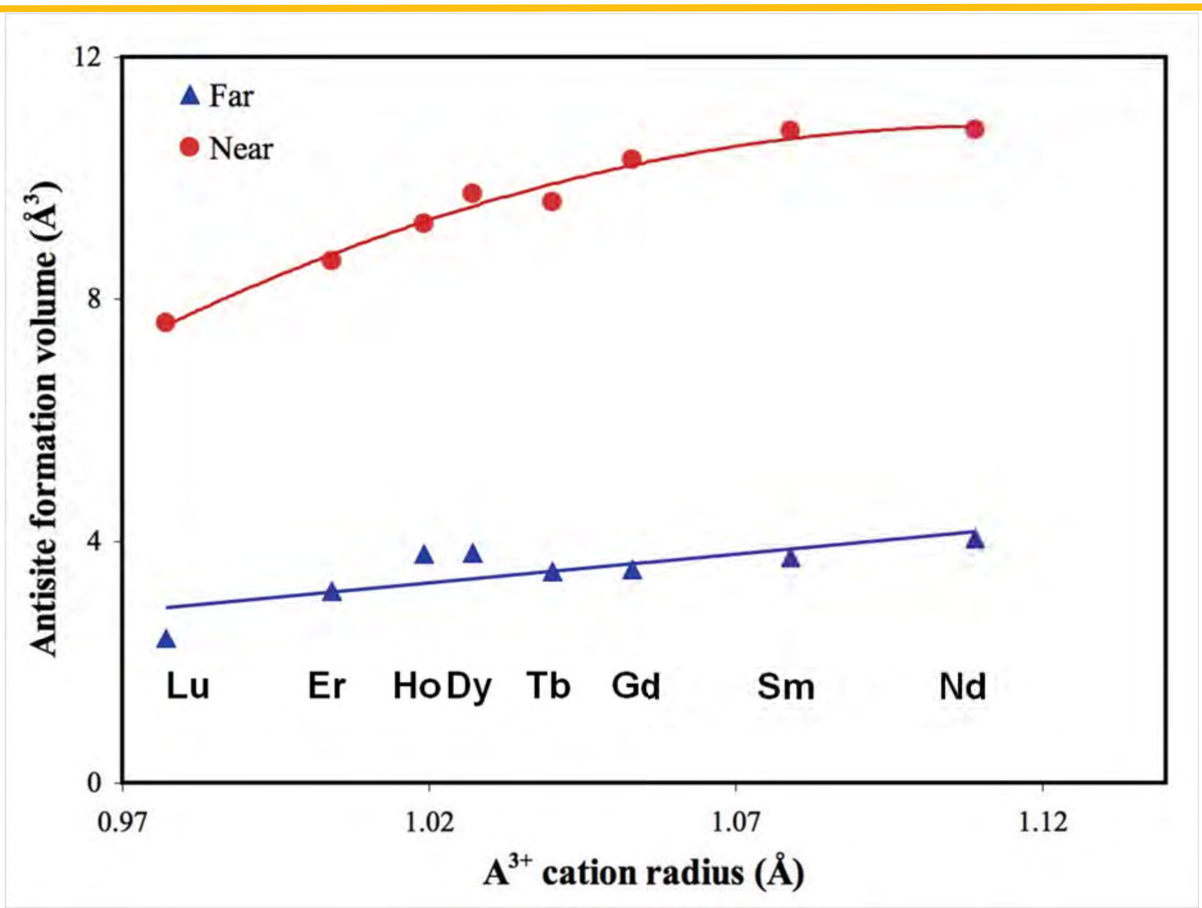
Cation Frenkel pair decay mechanism in $Lu_2Ti_2O_7$

(MD simulations by B. Uberuaga)



Antisite formation volume in $Ln_2Ti_2O_7$

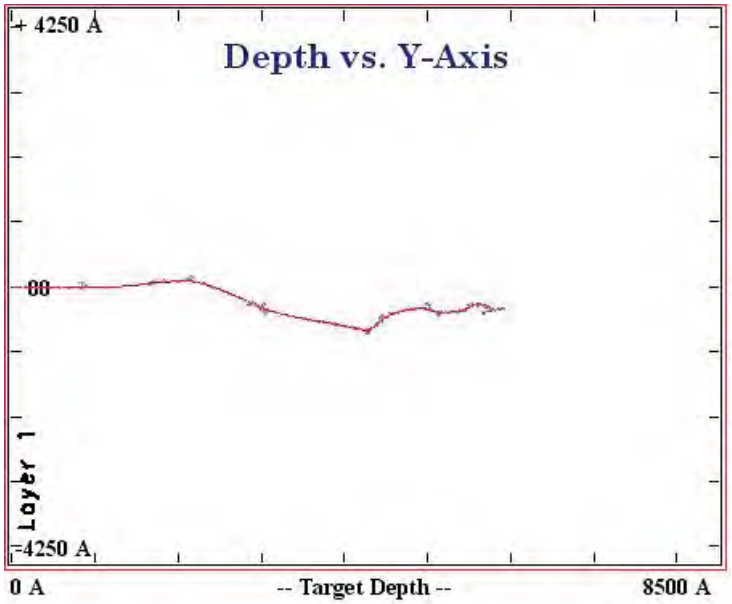
(DFT calculations by J. Chao)



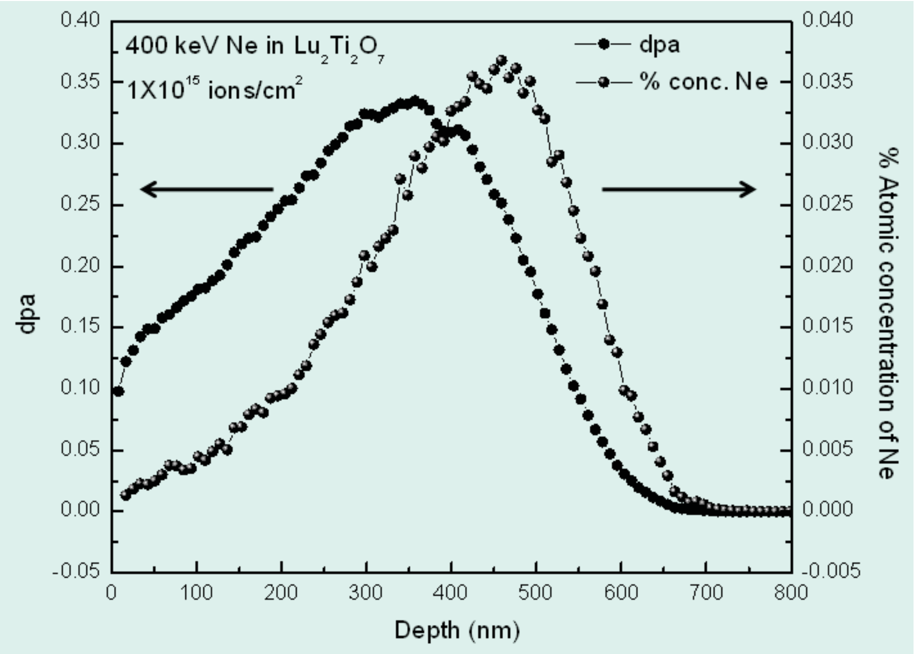
Design experiment to confirm hypothesis

$Lu_2Ti_2O_7$ pyrochlore irradiated at 77 K with 400 keV Ne^{++} ions

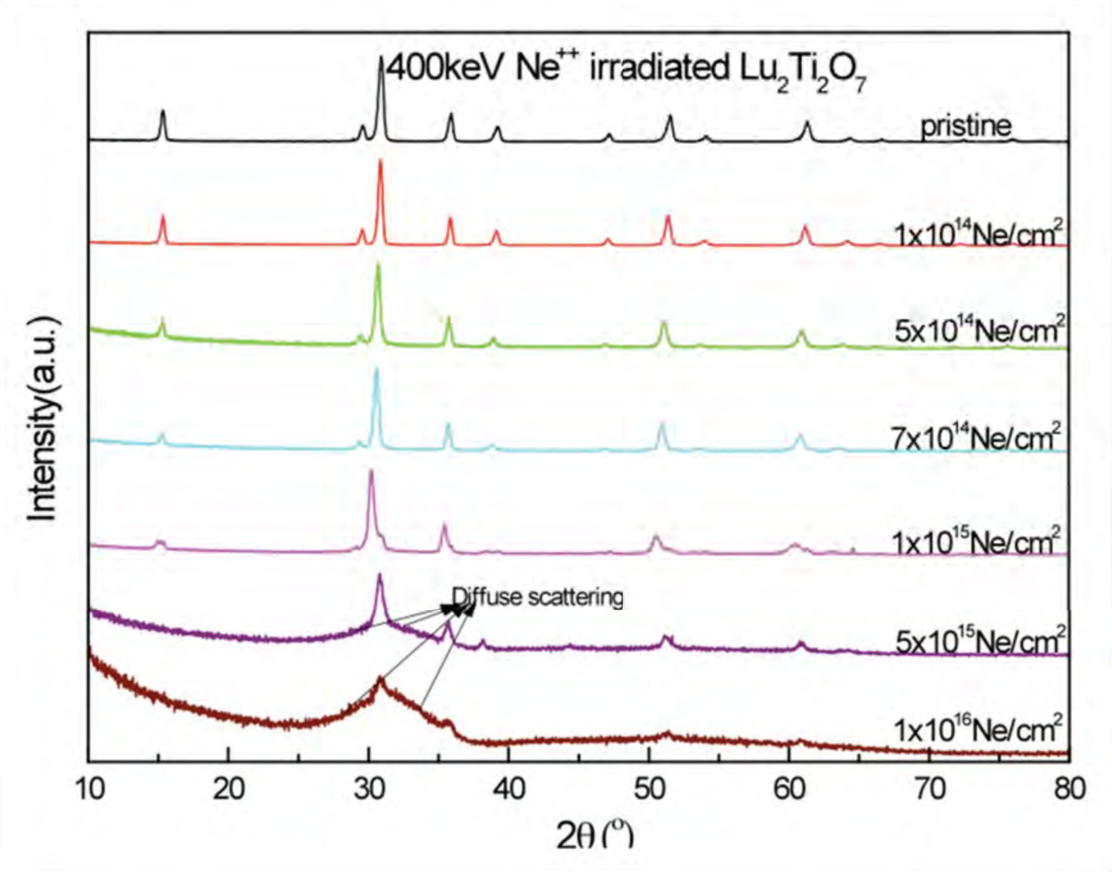
Single ion cascade morphology (from SRIM)



Displacements per atom (dpa) versus sample depth

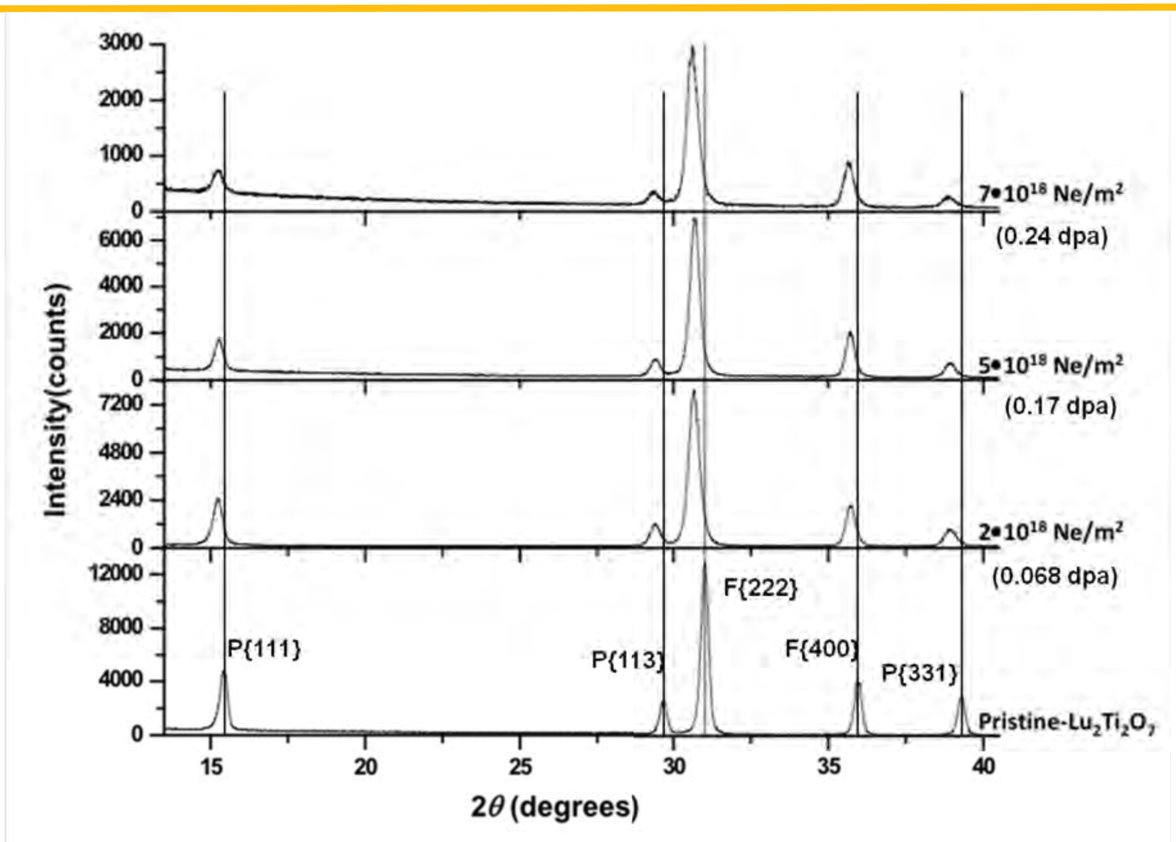


Grazing incidence XRD spectra versus dose
(γ = 0.25º, probing depth ~50 nm)



- 0 dpa
- 0.034 dpa
- 0.17 dpa
- 0.24 dpa
- 0.34 dpa
- 1.7 dpa
- 3.4 dpa

GIXRD data suggest lattice swelling and cation disordering in $Lu_2Ti_2O_7$ following 400 keV Ne^{2+} ion irradiation

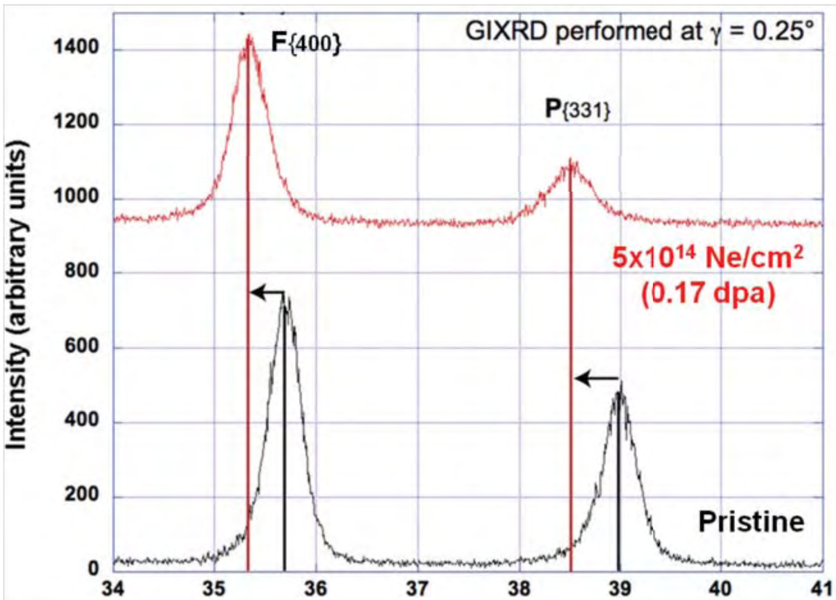


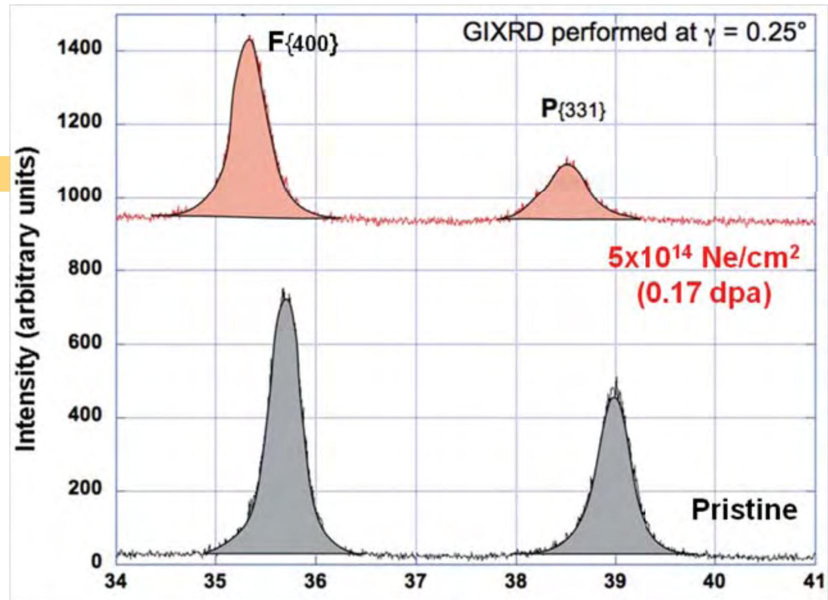
- ✓Interplanar lattice spacings increased with increasing the Ne fluence.
- ✓Pyrochlore super lattice peak intensity decreased with the Ne fluence.

Cation inversion (i): $Lu_2 Ti_2 O_7 \longrightarrow (Lu_{1-i} Ti_i)_2 (Ti_{1-i} Lu_i)_2 O_7$

i = 0; $(Lu_2)(Ti_2)O_7$, fully ordered   i=0.0625; one antisite formation

i = 0.5; $(Lu_{0.5}Ti_{0.5})_2 (Ti_{0.5} Lu_{0.5})_2 O_7$, fully disordered or amorphous state





Swelling (ΔV) ↔ Dose

$2d \sin\theta = \lambda$

$a = d_{hkl} (h^2+k^2+l^2)^{1/2}$

$\Delta V = V - V_0 = a^3 - a_0^3$

Inversion (i) ↔ I_P/I_F ↔ Dose

Peak intensity ~ Structure factor:

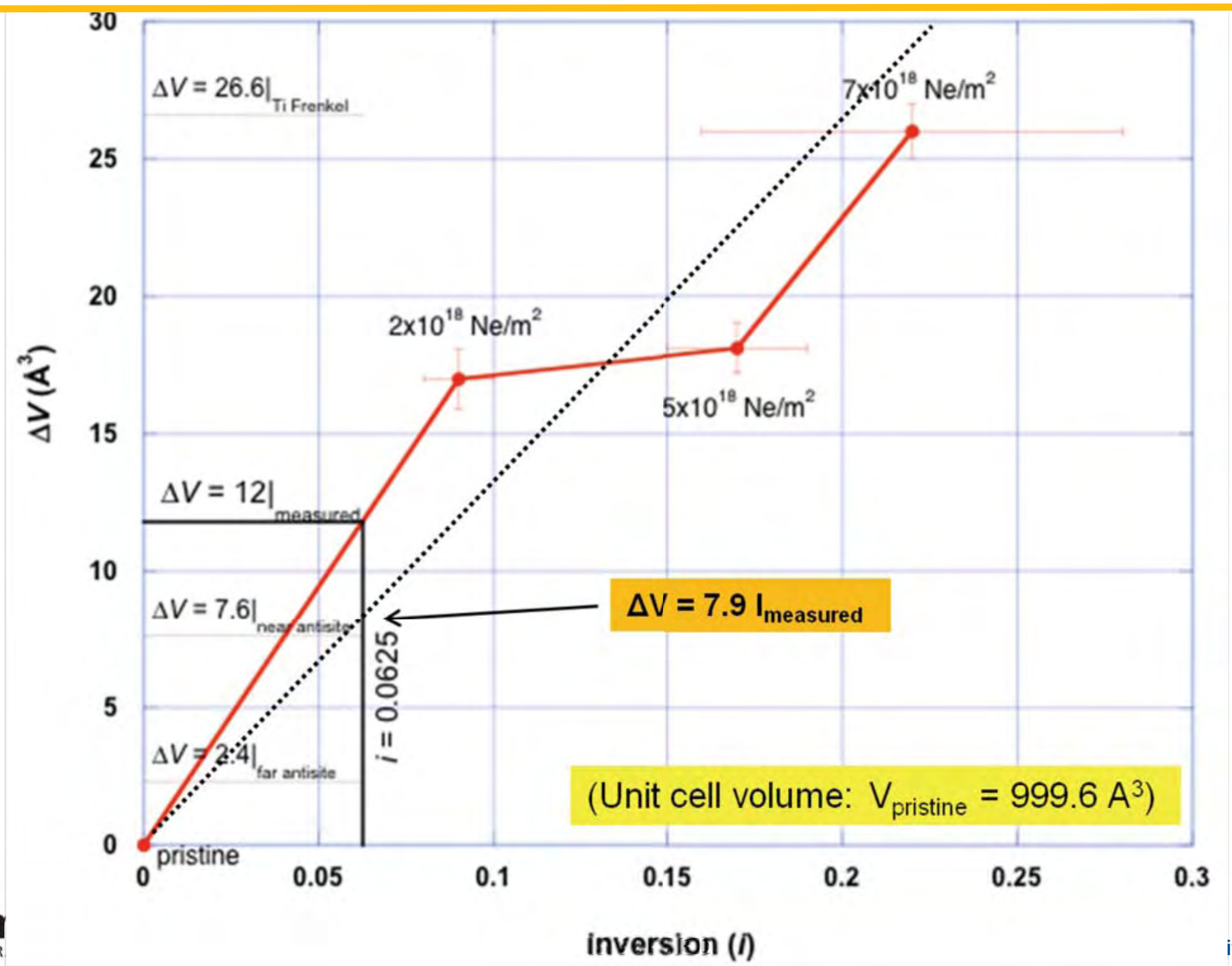
$\mathbf{I}_{331} \sim |F_{331}|^2$ and $\mathbf{I}_{400} \sim |F_{400}|^2$

Structure factor ~ Atomic form factor:

$F_{400} \sim f_{Lu} + f_{Ti}$ and $F_{331} \sim f_{Lu} - f_{Ti}$,

$\mathbf{I}_{331} / \mathbf{I}_{400} \sim \mathbf{i}$

Relationship of cation inversion and lattice swelling in $Lu_2Ti_2O_7$ following 400 keV Ne^{2+} ion irradiation



Summary of titanate pyrochlore radiation damage susceptibility

- ✓ First combined experiment and modeling approach to determine antisite volume in titanate pyrochlore structure
- ✓ Lanthanide titanate pyroclores (e.g. $Lu_2Ti_2O_7$) suffer volume swelling (1.7 - 2.5%) under low-temperature radiation damage conditions at doses of less than one dpa. They then succumb to an amorphization transformation at about 5% swelling.
- ✓ The swelling induced by irradiation is due primarily to cation antisite point defect formation and anion sublattice distortions that accompany the formation of these antisites.

Y.H. Li et al. under review at **PRL** (2011)

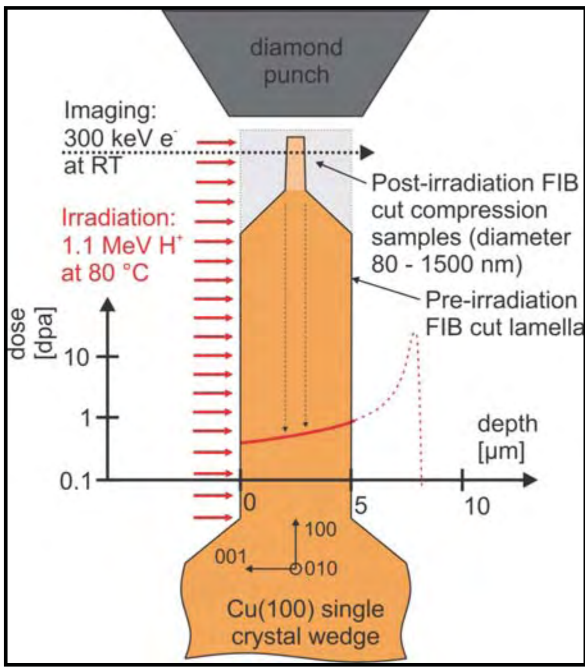
In situ nanomechanical testing on ion irradiated materials

A case study on single crystal Cu (100)

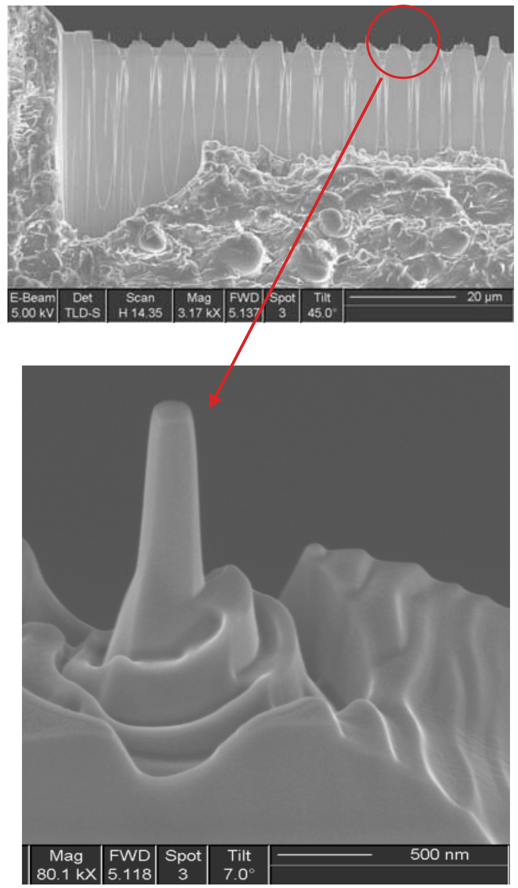
FCR&D (P. Hosemann and S. Maloy)

Motivation

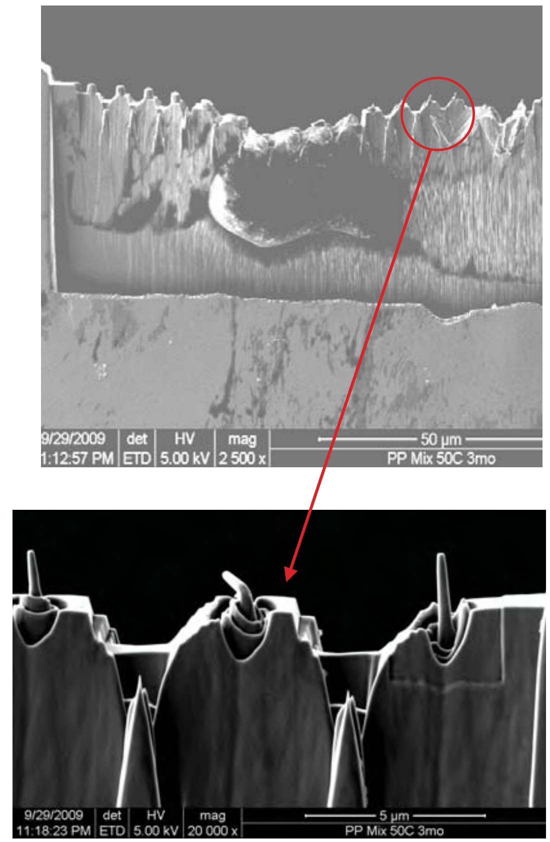
Is nanomechanical testing on ion irradiated samples a viable way to obtain bulk mechanical properties?



Before irradiation



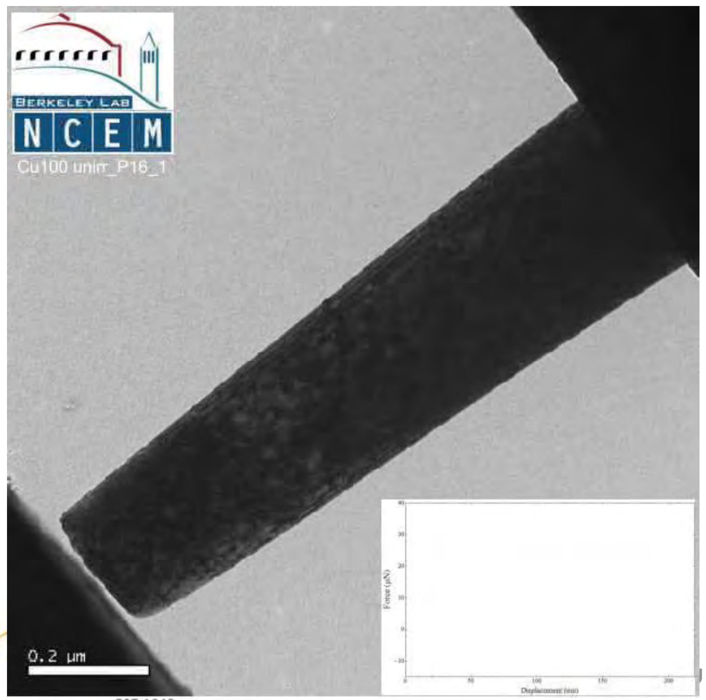
After irradiation



In situ compression testing on Cu (100) under TEM

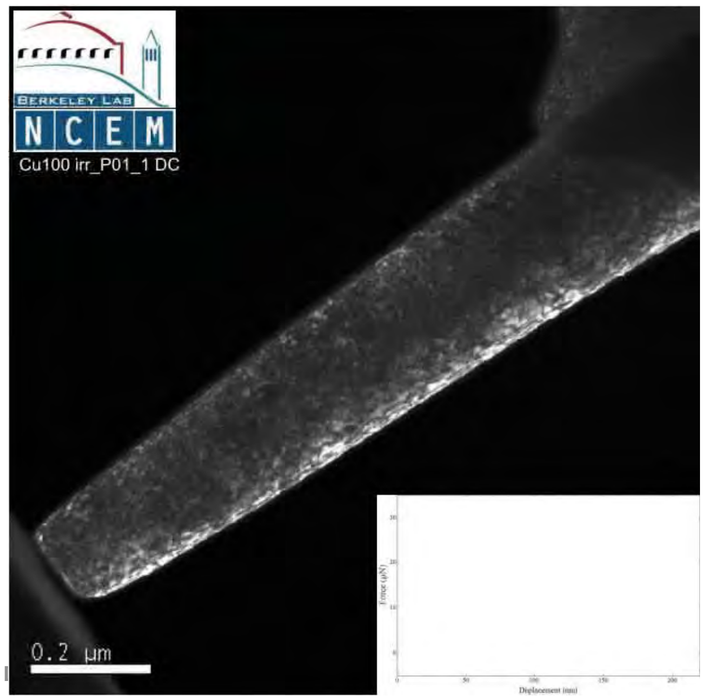
Before irradiation

Material: Cu (100)
Testing: Displacement controlled, 1 nm/s
Dimensions: d_{top}= 136 nm d_{avg}= 206 nm h= 904 nm



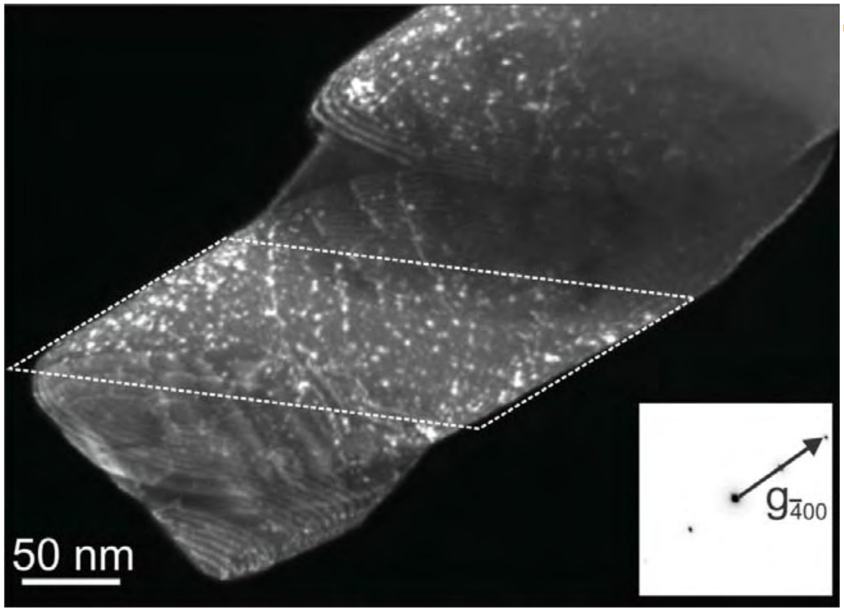
After irradiation

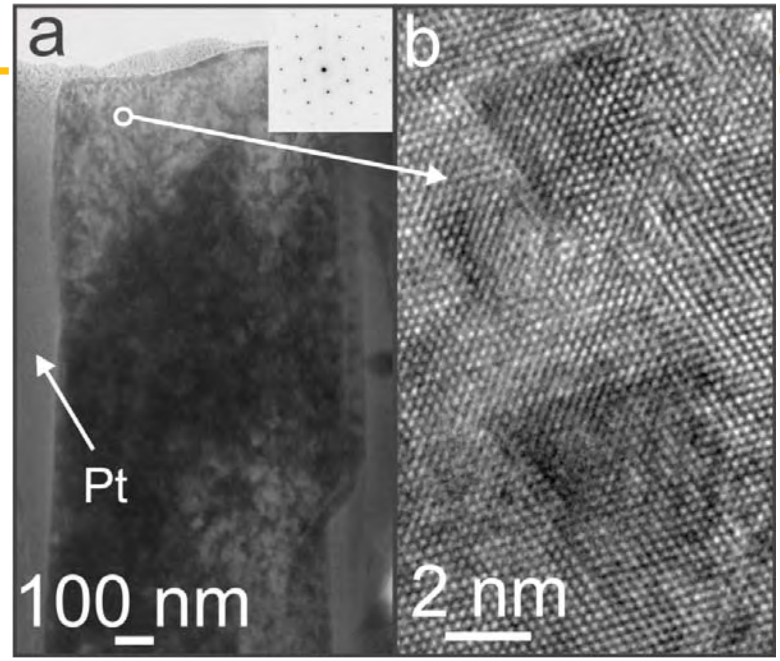
Material: Cu (100) irradiated to 0.8 dpa
Testing: displacement controlled, 1 nm/s
Dimensions: d_{top}= 118 nm d_{avg}= 198 nm h= 1143 nm



Post-compression TEM examination

Ion irradiated pillar structure

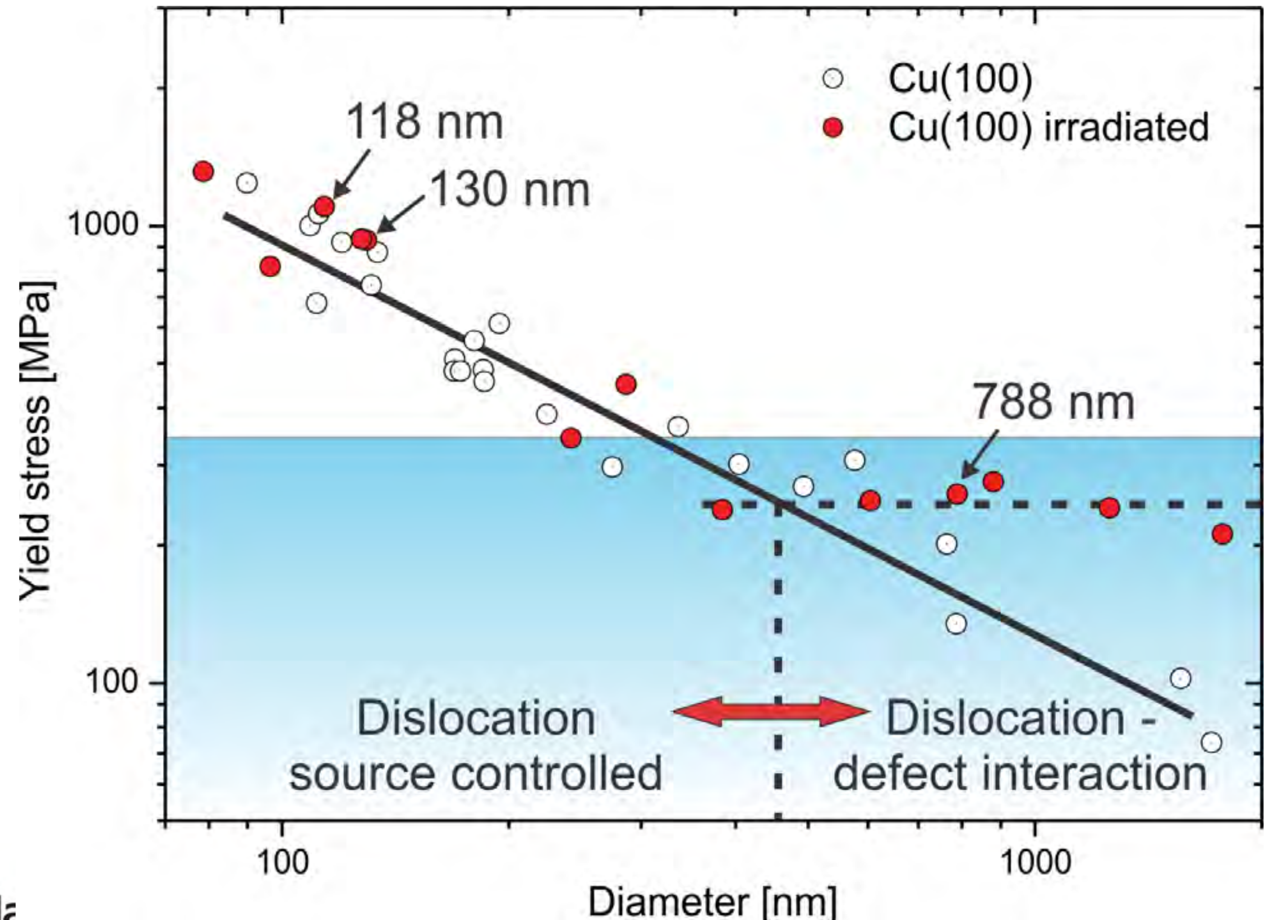




- ✓Slip localization on a slip plane in the middle of the sample
- ✓Many defects in the undeformed regions
- ✓ Defect densities: $1.4x10^{23}$ m^{-3}
- ✓ Defect spacing: ~20 nm

Stacking fault tetrahedrons (SFT) as pinning defect blocks

Size dependent of yield strength from micropillar compression testing



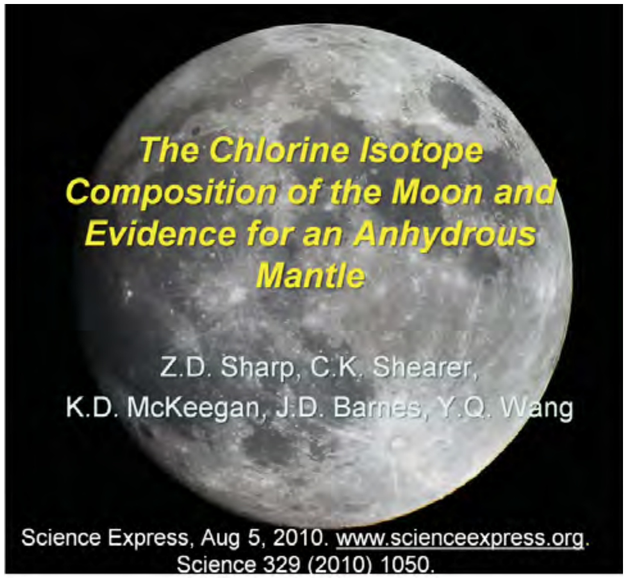
Summary of in situ nanomechanical testing of ion irradiated Cu(100) micropillars

- ✓ Quantitative in situ compression tests on irradiated samples (micropillars) under TEM observation
- ✓ Direct observation of radiation induced hardening and reduced ductility
- ✓ Bulk hardening was observed on samples as small as <u>450 nm</u> but not smaller
- ✓ Stacking fault tetrahedrons (SFT) as pinning defect blocks

<u>D. Kiener et al. under review at **Nature Materials** (2011)</u>

Mimic solar wind impact on Moon rocks:

Effect on $^{37}Cl/^{35}Cl$ ratios



- ✓ Chlorine is a volatile, hydrophilic element. On Earth it strongly tracks with water.
- ✓ Primitive mantle materials & carbonaceous chondrites are all close to 0‰.
- ✓ The isotopic composition of the Moon could indicate:
  - ✓ Similarity to Earth (no fractionation during Giant impact)
  - ✓ Differences due to:
    - – *impact fractionation?*
    - – *different source?*
    - – *late addition of volatiles to Earth*

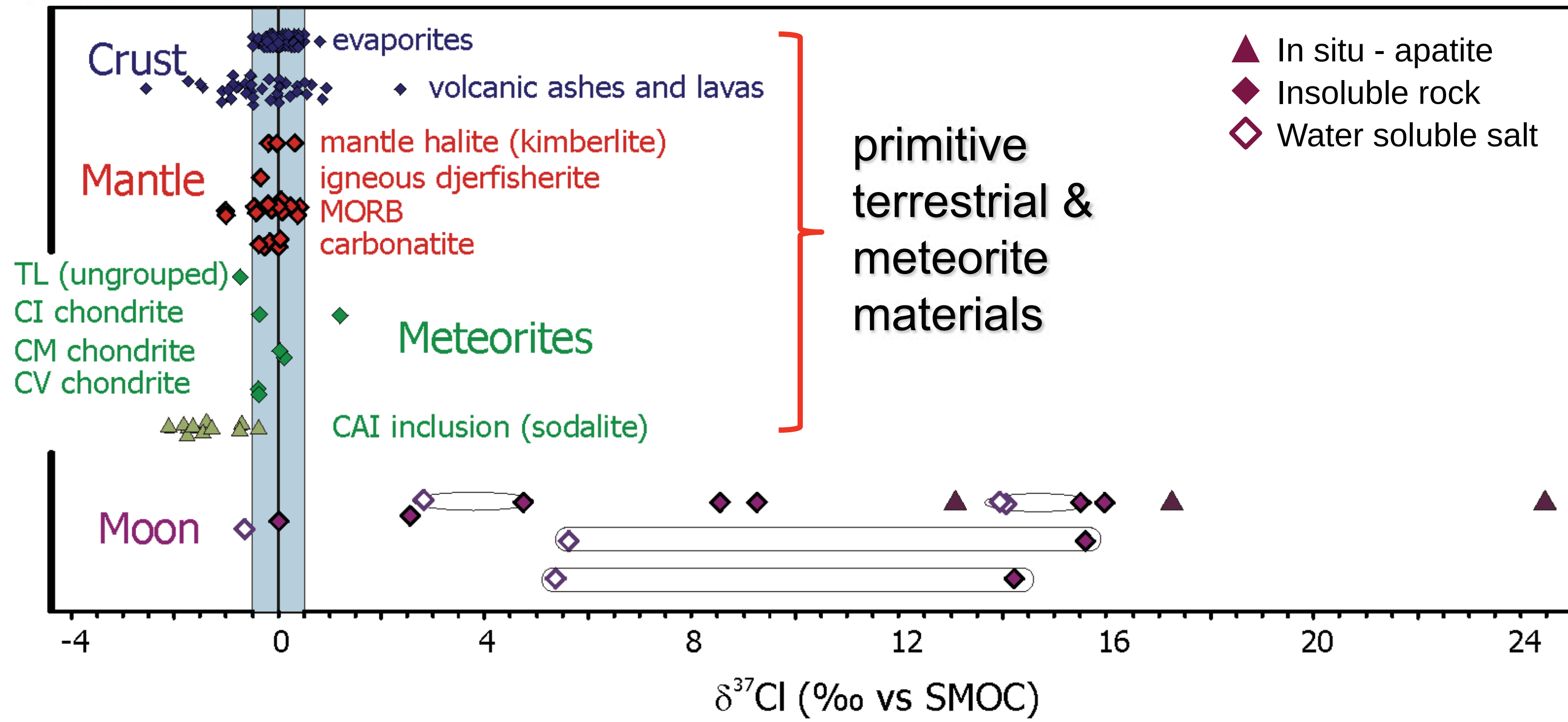
Chlorine Isotope Geochemistry

- ✓ Two stable isotopes of Cl: ^{35}Cl (75.8%); ^{37}Cl (24.2%) AMU = 35.45
- ✓ Data are given in conventional delta notation:

$$\delta^{37}Cl = \left(\left(\frac{^{37}Cl}{^{35}Cl}\right)_{sa} \Big/ \left(\frac{^{37}Cl}{^{35}Cl}\right)_{std} - 1\right)1000$$

- ✓ Standard is SMOC (*Standard Mean Ocean Chloride*)

Chlorine isotope composition



Why the spread?

1) ~~Fractionation during initial lunar heterogeneity~~
2) Fractionation during degassing
3) Fractionation due to solar wind bombardment

Fractionation during degassing of basalts

Earth

- **Cl degasses as HCl (*g*)**
- Two fractionation mechanisms occur:
  - *preferential loss of ^{35}Cl to vapor phase (higher translational velocity – Graham's Law)*
  - *preferential enrichment of ^{37}Cl in HCl (g) due to stronger bonding*
- Overall, the two effects cancel one another and <u>no Cl isotope fractionation is observed on Earth basalts</u>.

Moon

If Moon is anhydrous:

- Cl degasses as metal chlorides
- One fractionation mechanisms occurs:
  - *preferential loss of ^{35}Cl to vapor phase (Graham's Law)*
  - *No preferential enrichment of ^{37}Cl in metal chloride gases – similar bonding to melt*
- Degassing removes the light isotope of Cl, and results in <u>increased $\delta^{37}Cl$ values on Moon basalts</u>

Why the spread?

1) ~~**Fractionation during initial lunar heterogeneity**~~
2) **Fractionation during degassing (yes)**
3) **Fractionation due to solar wind bombardment (?)**

Does solar wind/micrometeorite bombardment cause Cl fractionation?

Design experiment to test hypothesis:

- ✓ Choose samples with different degrees of surface exposure
- ✓ Conduct high energy hydrogen bombardment study at IBML

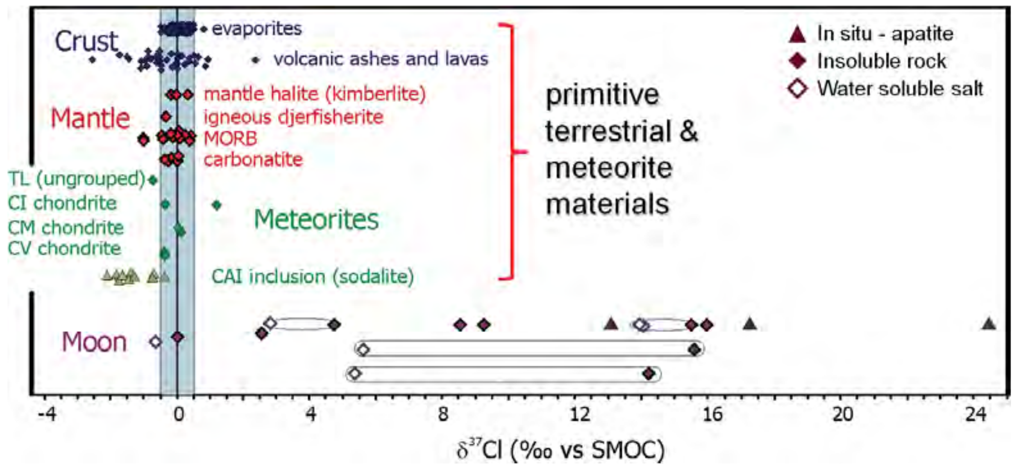
There is no correlation with degree of exposure and δ^{37}Cl value:

- ✓ The highest δ^{37}Cl value is for shielded apatite from KREEP pigeonite basalt 72275
- ✓ No correlation are observed with sulfur isotopes

IBML proton beam bombardment/irradiation:

- ✓ 2” diameter Al disk coated with NaCl film (2 µm thickness)
- ✓ the disk was irradiated with 100 keV protons to $1x10^{18}$ H/cm^2 at LN_2
- ✓ high resolution SIMS was then conducted to determine δ^{37}Cl value
- ✓ no meaningful increase was observed on the irradiated sample, when compared with the control sample.

Summary of mimic solar wind impact on Moon rocks



Why the spread?

1) ~~**Fractionation during initial lunar heterogeneity**~~
2) **Fractionation during degassing**
3) ~~**Fractionation due to solar wind bombardment**~~

The Moon is anhydrous (no water) !

Z.D. Sharp et al, **Science**, 329 (2010) 1050.

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- ✓ Understanding physics of palladium hydride behavior
- ✓ Pre-amorphization swelling in ion irradiated titanate pyrochlore oxides
- ✓ In situ nano-compression testing on ion irradiated Cu under TEM
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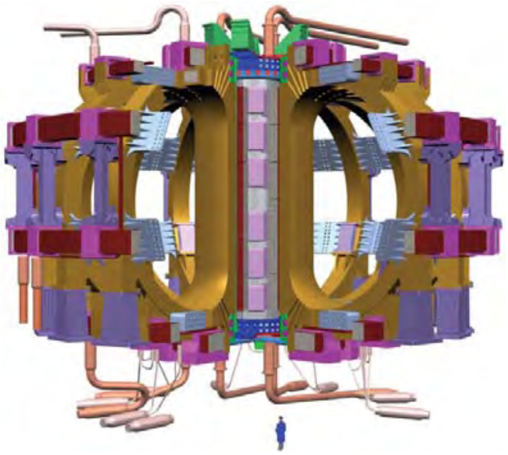
Future research plans

Ion Beam Materials Research

The need for radiation resistant materials

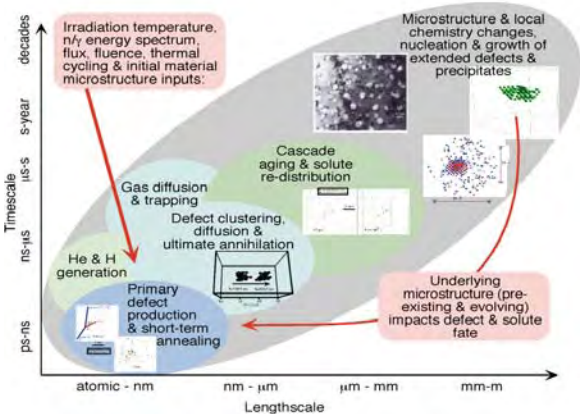
- ✓ **Nuclear fission and fusion reactors will require materials that can survive extreme environments of irradiation, temperature, stress, corrosion…**
- ✓ **Electronics for deep space missions must be rad-hard**

Nuclear Energy

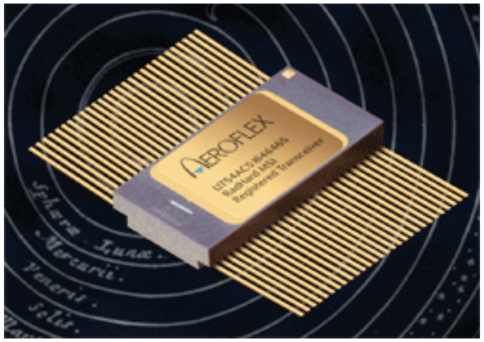


▪ITER website: www.iter.org

Radiation damage science



Rad-hard electronics



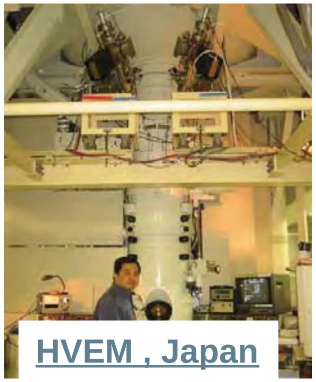
▪http://www.electronicproducts.com/

Novel materials synthesis under irradiation extremes, radiation damage, and in situ materials characterization

Frontier of materials radiation damage science

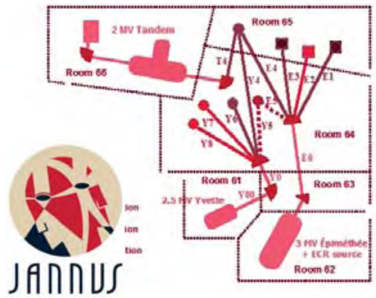
Radiation tolerant materials by design

Beyond "Cook and Look:" in situ diagnostics and tailored materials

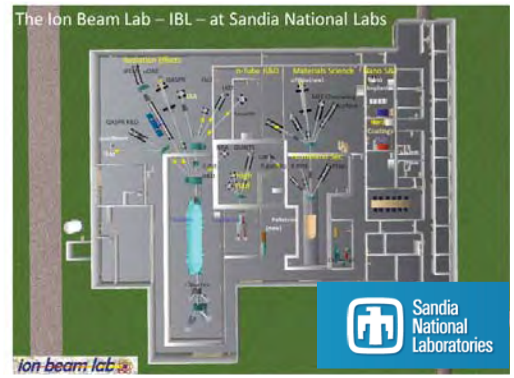


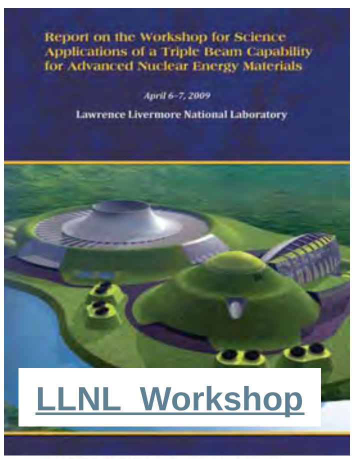
HVEM , Japan

CEA-Saclay Triple Ion Beam



IVEM , ANL





LLNL Workshop

M4 – IBML – F³

Los Alamos

Basic science question:

Use of ion implantation/irradiation to control microstructure and thus physical/ chemical properties of materials at the nanometric scale.

Synergistic effects between ballistic and electronic energy losses.

Nuclear energy relevance

Study of the irradiation behavior of nuclear materials and fuels considered for fission and fusion reactors

Synergistic effects between displacement damage and helium/hydrogen atoms in fusion/fission materials

T. Tanaka et al., Elsevier Science Bv., Kyoto, Japan, 2005, pp. 294.

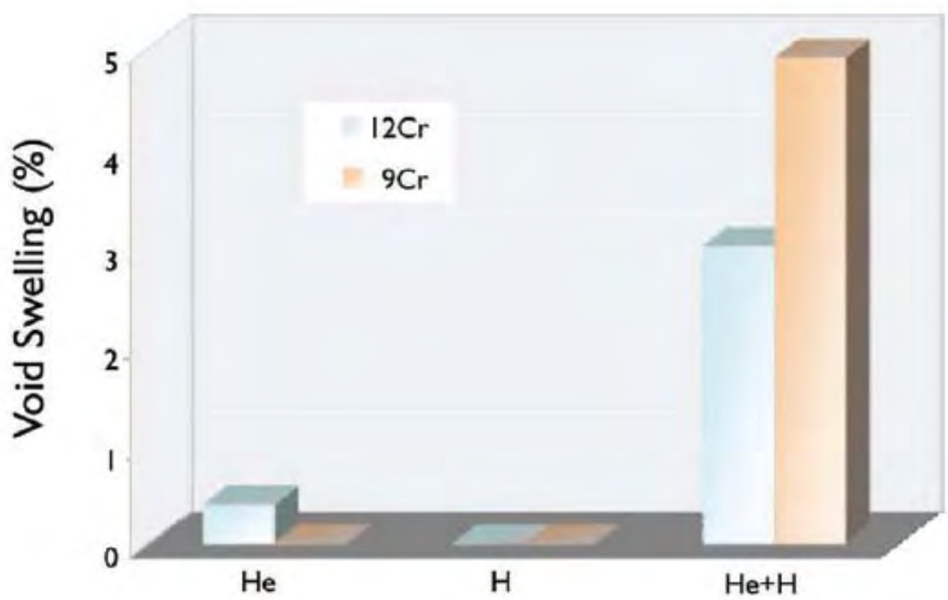


Figure 1. The synergistic effect of He and H was shown clearly in the triple ion (Fe^{3+} + He^{+} + H^{+}) irradiation of an FeCr steel.[3]

ELSEVIER

Nuclear Instruments and Methods in Physics Research B 99 (1995) 566–568

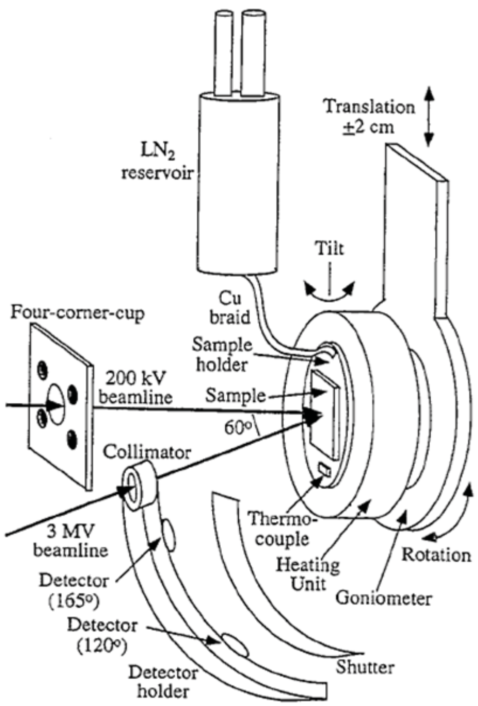
NIM B Beam Interactions with Materials & Atoms

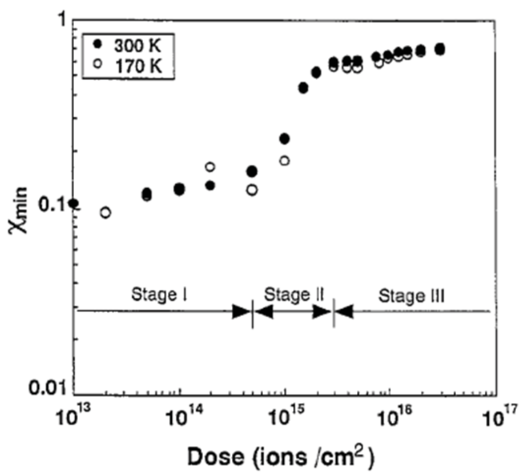
1995

In-situ capability of ion beam modification and characterization of materials at Los Alamos National Laboratory ☆

Ning Yu [a,*], Michael Nastasi [a], Timothy E. Levine [b], Joseph R. Tesmer [a], Mark G. Hollander [a], Caleb R. Evans [a], Carl J. Maggiore [a]

[a] Materials Science and Technology Division, Los Alamos National Laboratory, Los Alamos, NM 87545, USA
[b] Department of Materials Science and Engineering, Cornell University, Ithaca, NY, 14853, USA



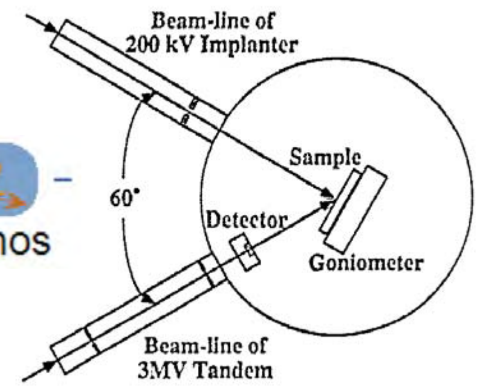


2011

Dual-beam irradiation

(still the only dual-beam setup in US)

IBML Los Alamos



Synergistic effect on HT-9:

300 keV He implant
2 MeV Fe irradiation

TEM characterization

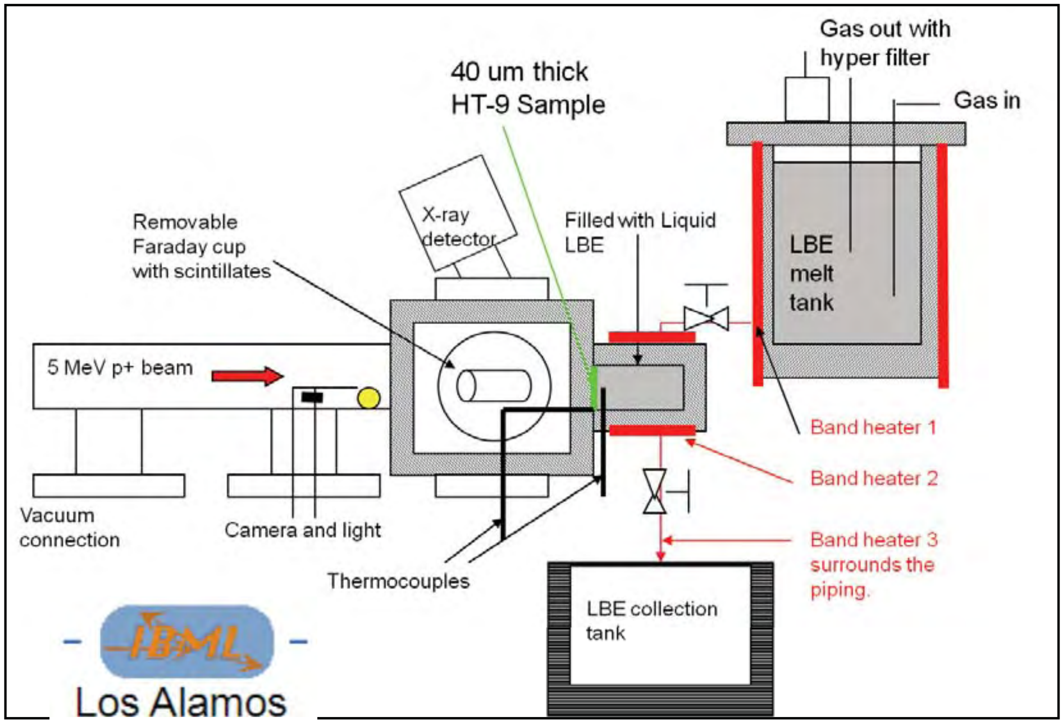
To compare with literature data

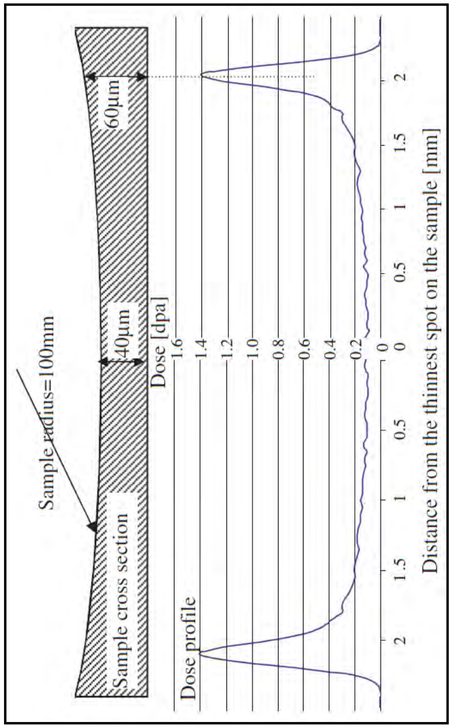
First ion irradiation and corrosion setup

Irradiation and corrosion experiment (ICE) at IBML (2008)

(P. Hosemann et al., JNM, 376 (**2008**) 392)

(HT-9 irradiated as exposing to liquid metal (lead bismuth eutectic) at 400ºC)

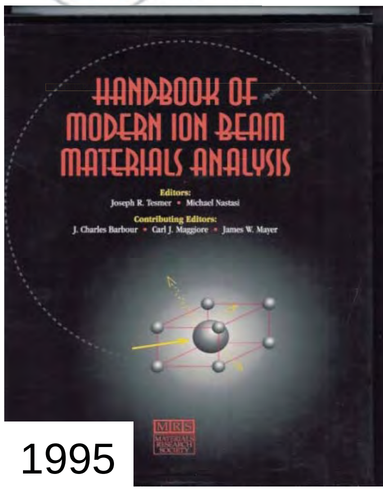




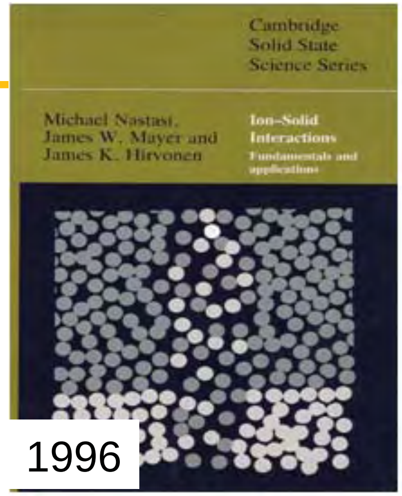
ICE-II at IBML (2011):

higher dpa (2~3 dpa), higher temp (600ºC), & better chemistry control

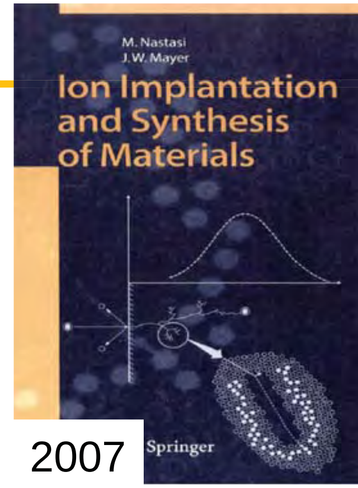
Ion beam materials research achievements at LANL
(1986-2011)



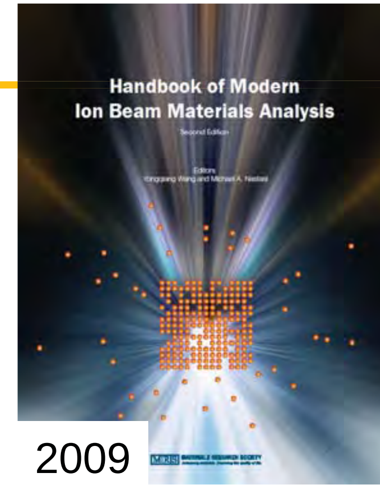
1995



1996



2007



2009

IBML related publications: Well over <u>**800**</u> refereed papers have been published by LANL researchers using the IBML Facility since its inception, including journals like *Science*, *Nature Materials*, *Physical Review Letters,* etc.

IBML user sponsored conferences: MRS symposium (1989), IBMM (1996), IIT (2002), IBA (2003), REI (2005), CAARI (2006-2012), IBA (2013).

IBML supported programs: BES (EFRC, CINT, Single Investigators), LDRD, NE (AFCI/GNEP/FCR&D), Weapons, Space Programs, WFO, etc.

(Courtesy of Nastasi, Sickafus, Tesmer, Maggiore, Maloy, and many others)

Irradiation environment in the proposed Materials Test Station

E. Pitcher (LANSCE-DO)

The proposed Materials Test Station, to be built at the Los Alamos Neutron Science Center (LANSCE), will use the high-power proton beam from the LANSCE accelerator to create an intense neutron irradiation environment for nuclear materials testing. The primary mission is to test advanced fuels and materials for fast reactor applications, including fuels bearing minor actinides, in support of the Department of Energy Office of Nuclear Energy's Fuel Cycle R&D program. Damage rates of up to 15 dpa per year in iron can be achieved within the fuel irradiation region. Not only can the MTS perform integral testing of fuel rodlets subjected to prototypic fast reactor conditions, it is also well suited for conducting separate effects experiments that are critically important to understanding the underlying processes that contribute to fuel aging, and ultimately fuel failure. Separate effects testing of the type than can be conducted in MTS can validate modeling efforts that are used to simulate fuel performance. In addition to fission reactor fuels and materials testing, the MTS is also well suited for testing materials for fusion reactor applications.

LANSCE: Nuclear physics and material science

J.L. Ullmann (LANSCE-NS)

LANSCE Division has an active program studying the physics of neutron-induced reactions for basic science and applications in stockpile stewardship, homeland security, and reactor physics. Many of these studies have a direct impact on materials properties. An example is the study of single-event effects in electronics circuits, and LANSCE has an active program where manufacturers and end-users of critical electronic circuits test the response of circuits in a neutron beam that mimics the cosmic-ray induced atmospheric neutron flux. Another example is the study of H and He gas production by neutron induced reactions on structural materials such as iron and titanium. There is also a major program run jointly by LANSCE and Chemistry Division to provide radioisotopes for the national and global user communities. The Isotope Production Facility needs to study materials in extreme radiation environments in order to make reliable isotope production target assemblies, and also studies nuclear reaction cross sections to develop production methods to meet new isotope needs.

The role of grain boundaries in modifying radiation damage evolution in simple metals

X. Bai (MST-8); A. Voter (T-1); R. Hoagland (MST-8); M. Nastasi (MPA-CINT); B. Uberuaga (MST-8)

It is well accepted that grain boundaries serve as effective sinks for radiation-induced defects (interstitials and vacancies). However, insight into the atomic-scale origin of this behavior is still lacking. In order to meet the demands of future nuclear applications, the origin of this enhanced radiation tolerance must be understood.

We use molecular dynamics, temperature accelerated dynamics, and molecular statics to study radiation damage phenomena near a variety of grain boundaries in Cu over three different temporal regimes: the short-time damage production phase of a collision cascade; the longer-time scales over which defect annihilation and aggregation occur; and the thermodynamic-limiting behavior of the system. We find that both the production and the subsequent annealing of the radiation-induced defects are modified significantly by the presence of the grain boundary. In particular, we identify a new mechanism by which interstitials efficiently annihilate vacancies, promoting enhanced defect recombination. We compare to previous experimental results and identify three regimes over which different thermally activated processes are active, resulting in different responses, both better and worse than large-grained counterparts, of the material to irradiation. Our results show that nanostructured materials have a very sensitive response to irradiation and offer new insights into the design of radiation tolerant materials.

Atomistic modeling and experimental studies of the shock response of Cu/Nb nanolayered composites

T.C. Germann (T-1); R.F. Zhang (T-3); J. Wang, X.-Y. Liu (MST-8); S.N. Luo (P-25); W.Z. Han (P-24); I.J. Beyerlein (T-3); A. Misra (MPA-CINT)

Classical molecular dynamics (MD) simulations, and laser and gas gun experiments are used to study the shock response of Cu-Nb nanolayered composites. Previous work has demonstrated the unusual radiation tolerance and mechanical strength of such materials due to the ability of Cu/Nb heterointerfaces to absorb both point defects (thus catalyzing Frenkel pair recombination) and line defects (by delocalizing them at the easily sheared interfaces), respectively. Initial laser experiments and MD simulations indicate that shock-induced defects in Cu/Nb nanolaminates are largely recovered upon unloading due to the confinement of plasticity (slip or twinning) within the layers, and the ability of interfaces to absorb dislocations.

Here we describe the development of an embedded atom method (EAM) interatomic potential, which provides an accurate description of deformation twinning in bcc Nb under compression, slip in fcc Cu, and the interface structure of Cu-Nb interfaces with the Kurdjumov-Sachs (KS) orientation relationship. Using this potential, MD simulations provide insight into the role of atomic Cu-Nb interface structures on the nucleation, transmission, absorption, and storage of dislocations during shock loading. The key role which interface structure plays is demonstrated by post-mortem transmission electron microscopy (TEM) of shock-recovered samples, which indicate that deformation twinning in Cu is preferentially nucleated from Cu(112)//Nb(112) interface habit planes. Corresponding MD simulations are underway.

Irradiation damage effects in ceramic oxides induced by high temperature and high fluence ion beam irradiation

I. Usov, D. Devlin (MST-7); J. Won, M. Hawley (MST-8); A. Suvorova (University of Western Australia); Y. Wang, K. Sickafus (MST-8)

This work is motivated by our interest in understanding the mechanisms responsible for irradiation-induced defects nucleation, growth and recovery, and our goal to discover ceramic oxides resistant to irradiation. Radiation tolerant ceramic oxides are considered to be attractive for various nuclear energy applications (for both fission and fusion type reactors). An important criterion for selection of suitable materials is their tolerance to a combination of irradiation-induced damage and elevated temperature. A major cause of radiation damage in nuclear reactor components is the stopping of fast neutrons, fission fragments, high-energy alpha particles and recoil nuclei formed following alpha decay. To simulate a nuclear reactor radiation environment, we have employed ion beam irradiation. A common feature of the ion irradiation is that ions can penetrate only to a particular depth and therefore nature of ion irradiation-induced defects substantially depends on the surface proximity. In this poster we'll present an experimental study of ion irradiation-induced effects (surface blistering, gas filled bubbles formation, amorphization and lattice damage recovery) in various ceramic oxides. Particular attention will be given to the high dose and high temperature effects on modification of the near surface and bulk properties.

This work was supported by the U.S. Department of Energy (DOE) Office of Basic Energy Sciences, Division of Materials Sciences; the DOE Advanced Fuel Cycle Campaign and Fuel Cycle R&D Program; and the Los Alamos National Laboratory's Directed Research and Development (LDRD DR) grant.

IBML proton calibration results of the SABRS Energetic Particle Subsystem (ZEP FU1)

J. Burward-Hoy (ISR-1) and the ZEP Team: K. Grace, S. Hahn, J. Battles, J. Archer, S.D. Salazar, R. Ortiz, A. Gonzales (ISR-4); and MST Team: Y. Wang, J. Tesmer, R. Greco (MST-8), I. Usov (MST-7)

The energetic particle instrument (called ZEP) of the Space and Atmospheric Burst Reporting System (SABRS) was calibrated in 2010 by teams from both ISR and MST divisions. The ZEP measures both protons and electrons with different pitch angle distributions over a wide energy range (50 keV-70 MeV) with its multiple look directions optimized in position for a three-axis stabilized host platform flying in a geosynchronous orbit. Particle identification is obtained by using a fast coincidence between front-end and back-end sensors and the E-dE/dx measurement technique. Scattering backgrounds are minimized with collimators comprised of high-Z plates. The instrument includes both high purity silicon transmission mounted detectors of varying thicknesses and a shared, fast YSO scintillator with a photodiode readout. SABRS is a satellite-based sensor suite deployed in collaboration with the United States Air Force to verify the Limited Test Ban Treaty and provide space weather data to national customers. SABRS represents the next generation in a series of satellite instruments developed at LANL for these purposes dating back to the early 1960's and the Vela Hotel program.

The MST team at the Ion Beam Materials Laboratory (IBML) provided proton energies that ranged from 0.5 MeV - 25 MeV in an experimental set-up that consisted of a scattering chamber with multiple port positions, and a beam line that adjoined a 3MV tandem accelerator and 200 kV ion implanter. To provide protons in this energy range, different beams, target foils, and port configurations in the scattering chamber were used: (1) a proton beam and Rutherford scattering off of a 50nm thick Au-foil kept count rates in the measurement range for the ZEP instrument for energies 0.5-6 MeV; and, (2) a 3He beam and an ErD2 foil induced nuclear reaction produced protons at 12.9 MeV at the 135º port with 90 p/uC/msr, 14.8 MeV at 90º port with 140 p/uC/msr, and 17.367 MeV, 21.47 MeV, and 25.236 MeV at the 30º port using 3He beam energies of 1, 4.5, and 8.6 MeV. Al-foil energy attenuations resulted in beam energies between 6-14.8 MeV. The ZEP response to protons in this energy range will be presented.

Effects of radiation environments on long-term aging of weapon materials (U)

J. Lillard (MST-6)

Weapon materials are subjected to both internal and external radiation sources during their lifetime. Integrated efforts across the Los Alamos National Laboratory have lead to a better understanding of dose, dose rate, and thermal effects on materials of interest and, in particular, some of the material properties of importance. The results of these efforts have improved our predictive models for stockpile stewardship.

Advanced materials development and testing for Fuel Cycle Research and Development program

O. Anderoglu (MST-8); J. Van den Bosch (SCK-CEN, Belgium); T. Saleh (MST-16); B.H. Sencer (Idaho National Laboratory); P. Dickerson, R. Dickerson, (MST-6); P. Hosemann (UC Berkeley); G.R. Odette, N. Cunningham, Y. Wu (UC Santa Barbara); D. Hoelzer (Oak Ridge National Laboratory); B. Wirth (University of Tennessee); A. Misra, M. Nastasi (MPA-CINT); S. Maloy (SPO-CNP)

One of the main objectives of the fuel cycle research and development program is to develop core materials that can withstand irradiation damage up to or greater than 400 dpa. This will facilitate high burn up of the fuel ($\geq$ 40%), which in turn reduces the waste while producing zero carbon emission electricity.

The materials under study include nanostructured ferritic alloys (NFAs) (e.g. 14YWT), Fe based model alloys, and ferritic/martensitic (F/M) alloys (e.g. HT-9 and T91). F/M alloys were previously irradiated in the fast flux test facility (FFTF) reactor to doses up to 184 dpa at irradiation temperatures from 380-546 ºC. This also includes analysis of a duct made of HT-9 after irradiation to a total dose of 155 dpa at temperatures from 410-470 ºC with lower dose material covering irradiation temperatures from 370-510 ºC.

In addition to optimization of the microstructure in large heats, the comprehensive study on NFAs includes initial ion beam irradiations, processing studies to manufacture thin walled tubing for cladding and appropriate welding techniques for joining.

As a complement to engineering studies on NFAs, research is underway on model alloys focusing on the crystal structure at the atomic level and phase and interface stability under irradiation of the oxide-metal interfaces. The studies on the advanced materials utilize advanced characterization techniques including transmission electron microscopy (TEM), atom probe tomography (LEAP), and small angle neutron scattering (SANS).

Influence of interstitials or vacancies on grain boundary sliding processes in bcc tungsten

V. Borovikov, D. Perez, X. Tang (T-1); X.-M. Bai, B.P Uberuaga (MST-8); A.F. Voter (T-1)

Atomistic computer simulations were performed to study the influence of radiation-induced damage on grain boundary (GB) sliding process in bcc tungsten (W). For a number of GBs, we found the surprising result that introducing interstitials or vacancies into GB can reduce the average sliding-friction force under shear by more than an order of magnitude. Moreover, because these GBs typically shear in a well-known “coupled” way, we speculate that this may provide W with a built-in "self-healing” capability under irradiation conditions, as follows. A collision cascade produces vacancies and highly mobile interstitials; the diffusing interstitials find, and are trapped at, a nearby GB. The interstitial-loaded GB is now so easy to shear that internal stresses in the crystal may start it moving, and the coupled motion causes it to sweep past the cascade center, sweeping up the vacancies as it goes. We have also observed that as the number of interstitials in the GB is varied, the direction of the coupled motion sometimes reverses, causing the GB to sweep in the opposite direction under the same applied shear stress.

Overview: Materials for nuclear energy (focus on actinides)

D.F. Teter (MST-DO)

This talk will present a brief overview of materials research and applications for nuclear energy with a focus on actinide materials. Within this topic we will present technical highlights and accomplishments in the following three theme areas: metal fuels, ceramic fuels and waste forms, and non-proliferation. These theme areas illustrate a good synergy between modeling and experimentation as well as spanning synthesis, characterization and material performance. The connection of research areas with LANL's missions will be discussed as well as providing some metrics used for benchmarking. A review of the program portfolio will show the diversity of programs and how they synergistically help achieve LANL's mission in Materials for Nuclear Energy by translating science to solutions.

Overview: Materials for Nuclear Energy (Focus on Actinides)

David F. Teter

MST Deputy Division Leader

Materials for Nuclear Energy is a Good Illustration of the LANL Materials Strategy in Action

<u>Science</u>

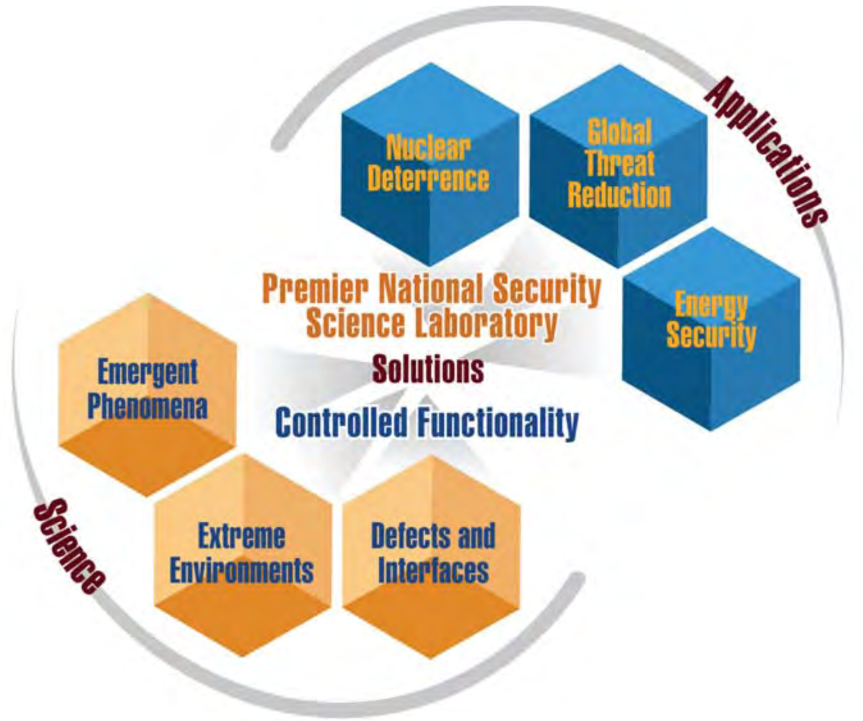
(Defects and Interfaces)

<u>Solutions</u>

(New Materials:
Fission gas management,
Radiation tolerance)

<u>Applications</u>

(Advanced Nuclear Energy
Fuel Forms)



LANL's Materials Strategy and Nuclear Energy Strategy are Consistent

LANL Nuclear Energy Strategy

Provide global leadership in Nuclear Energy Science to address the important science challenges for the continued development and safe implementation of sustainable nuclear energy.

Integrate experiment, theory, and modeling to develop practical solutions

Develop and demonstrate science based solutions for **getting the most from our existing nuclear energy infrastructure**.

Develop the **science and technology for future nuclear energy fuel cycle options**.

Identify potential risks, key points of impact, and **solutions for global nuclear materials management to address the current and evolving fuel cycle**.

Build and apply modeling and simulation capabilities for **enhancing and accelerating science and technology development and providing concrete information and outcomes for decisions regarding nuclear energy system options**.

Sustain and develop the required expertise, facilities, and partnerships

Build on key areas of strength: nuclear reactor fuels and materials , safeguards and non-proliferation , waste management and repository science, nuclear reactor design

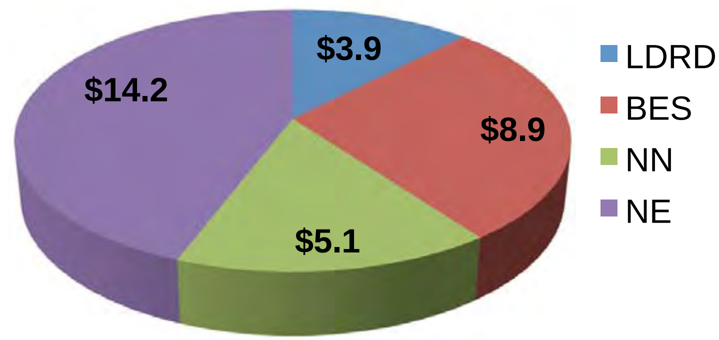
Materials for Nuclear Energy: Focus on Actinides has Three Main Theme Areas

- **Metal Fuels**
  - More application driven, process development
  - Alloy phase segregation, Shrinkage and Distortion, Interfacial reactions
- **Ceramic Fuels and Waste Forms**
  - Balance between science and application driven
  - Fundamental radiation damage phenomena and thermophysical properties
  - Phase Stability
  - Fission gas management
- **Non-proliferation**
  - More application / national policy driven
  - Material attractiveness
  - Materials development for advanced nuclear material detection

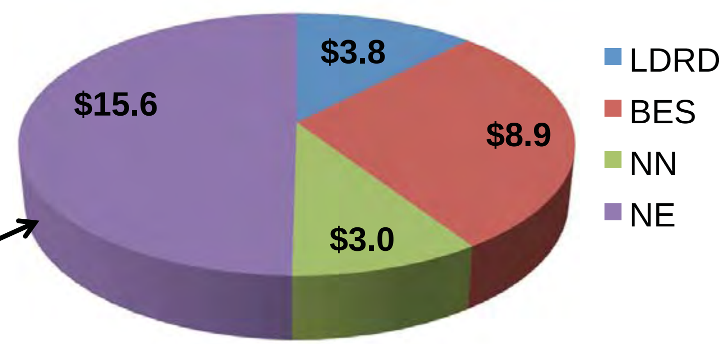
Diverse and Growing Materials Science Program in Nuclear Energy Portfolio

- **Nuclear Energy (NE) / Fuel Cycle R&D**
  - Advanced Fuel Development
  - Modeling & Simulation
  - Separation Research & Development
  - Waste Forms
- **LDRD**
- **Nuclear Non-Proliferation**
  - CONVERT
- **BES**
  - Core
  - EFRC

FY10 ($32.1 M)

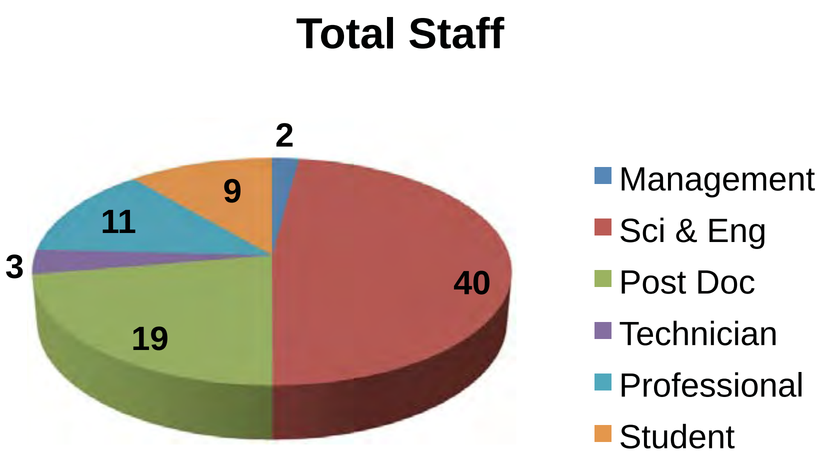


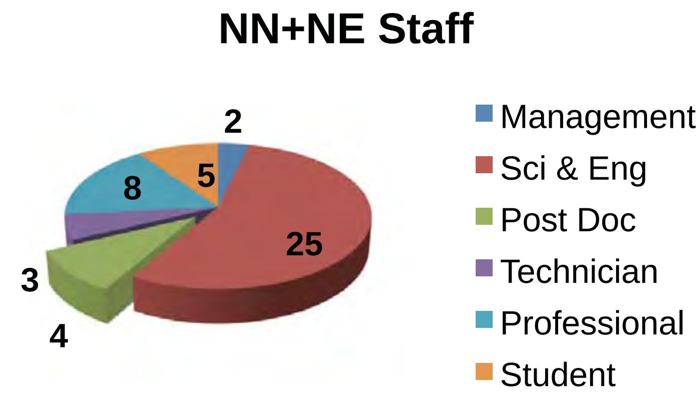
FY11 ($31.3 M)

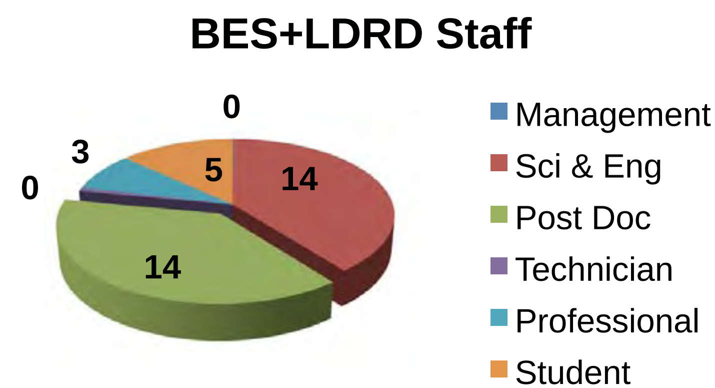


+$2.5M after CR

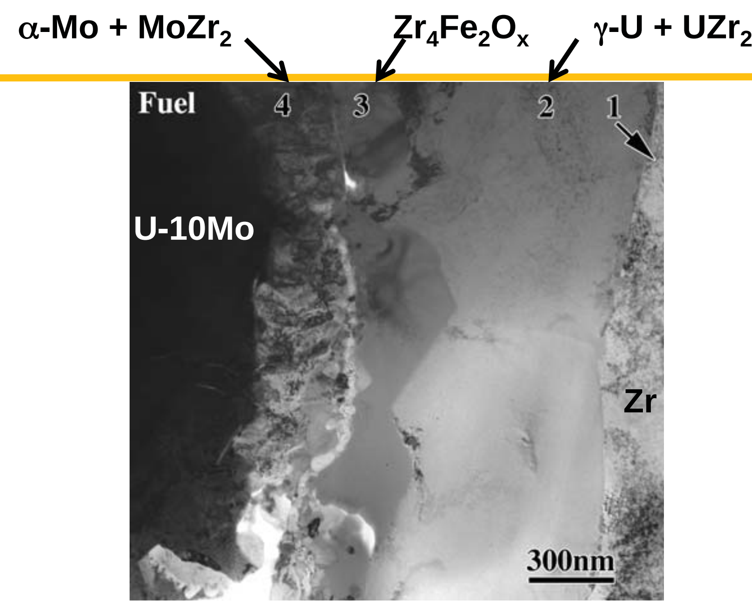
Materials Science Staffing Is Balanced Within The NE-related Programs With A Healthy Fraction Of Early Career And Post-docs



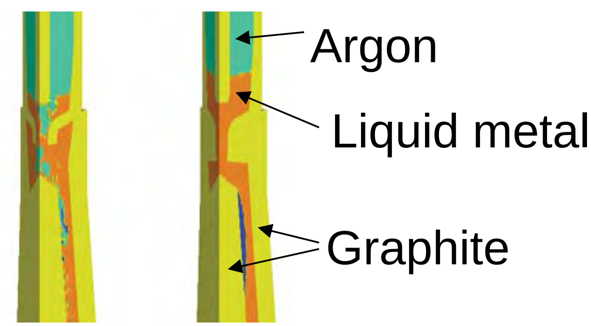




LANL is Using Expertise Developed for Nuclear Weapons Program for Development of Metal Fuels



- **CONVERT**
  - Performed rolling process development over 4 mo. timeframe
  - Coupled process modeling and experimental effort
  - Produced 6 Prototype U-10%Mo LEU fuel foils for AFIP-7 test at ATR
  - Performed state-of-art characterization on fuel plates produced at INL and LANL
- **Metal Fuel Modeling**
  - Performed Truchas-based modeling of actinide metal fuel casting operations at INL to guide process improvements.



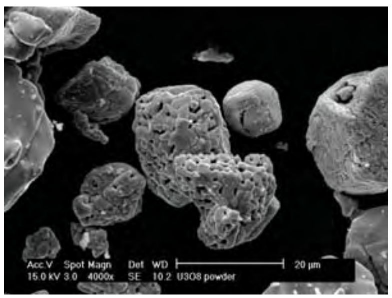
Dombrowski and Korzekwa (MST-6)

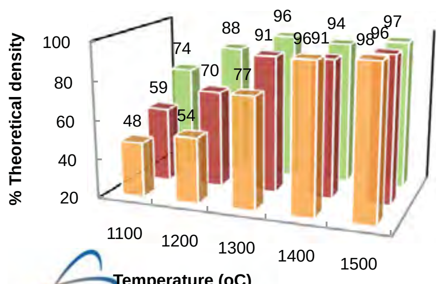
Simulations of argon gas driven filling, showing two different inlet geometries

Ceramic Fuel Development: R&D approach establishing and understanding the relationships between processing, structure, properties and performance

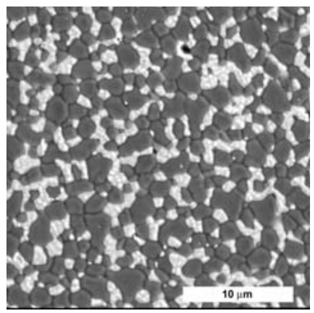
Understanding these relationships enables design and optimization of fuels, and when coupled with corresponding physics-based models, will enable prediction of subsequent performance

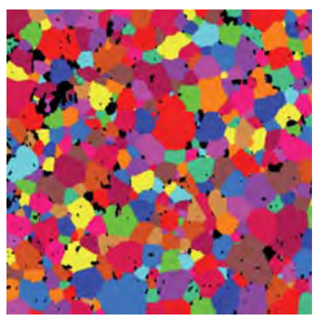
Processing



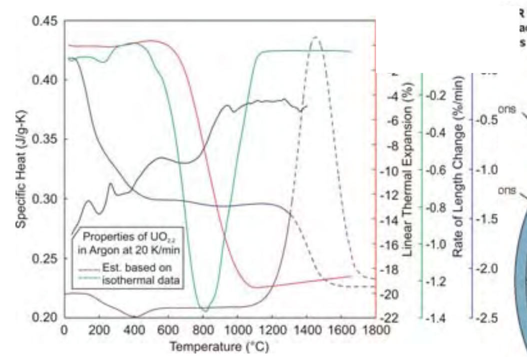


Structure

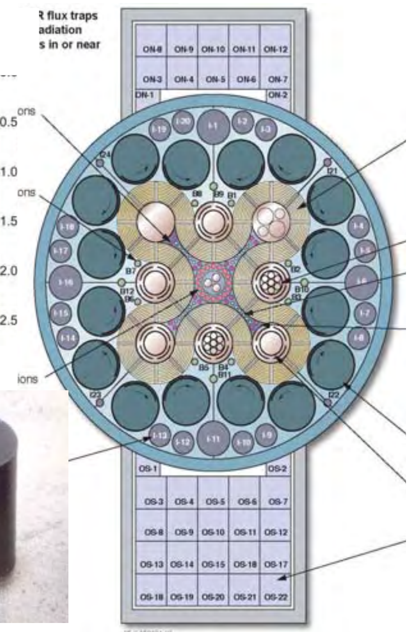




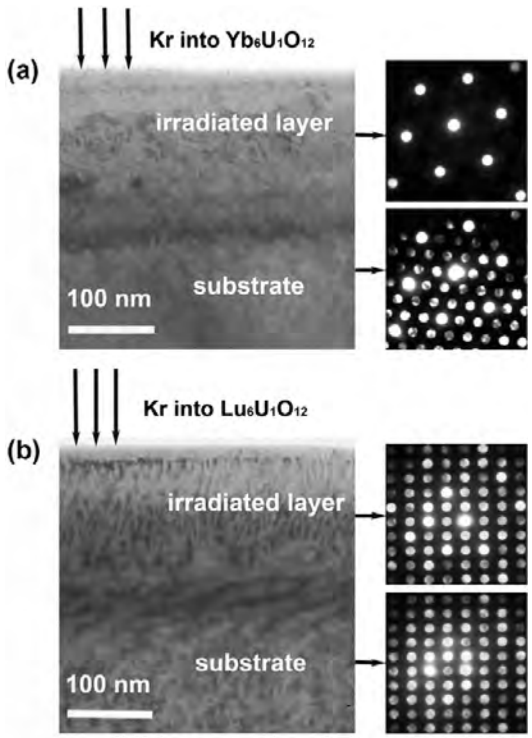
T_m,C_p,κ, λ, C_{ij}, etc



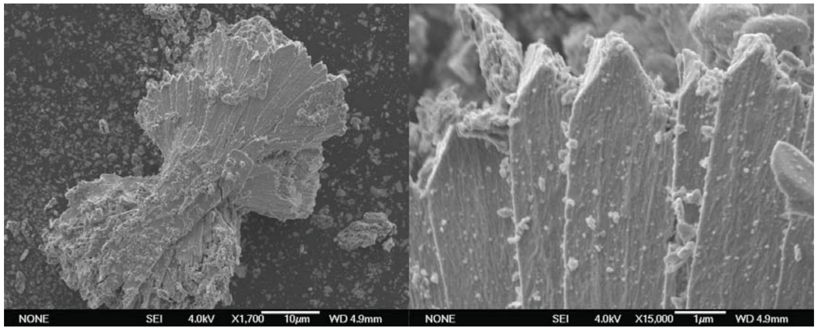
In-pile Performance



Ceramic Fuels and Waste Forms Highlights (Experimental)



PuO2 Precursor powder for MOX fuel Pellet



- A.D. Neuman et al., *Characterization of minor actinide mixed oxide fuel*, <u>Journal of Nuclear Materials</u> 385 (2009) 168–172
- Wheeler, K. et al., *Effect of Sintering Conditions on the Microstructure and Mechanical Properties of ZrN as a Surrogate for Actinide Nitride Fuels,* <u>J. Nuclear Materials</u>, 366 (2007) 306-316
- Tang M, et al. *Microstructural evolution in irradiated uranium-bearing delta-phase oxides A(6)U(1)O(12) (A = Y, Gd, Ho, Yb, and Lu),* <u>Journal of Nuclear Materials</u> 407 (2010) 44-47
- Sickafus KE, et al., *Radiation-induced amorphization resistance and radiation tolerance in structurally related oxides*, <u>Nature Materials</u> 6 (2007) 217-223. **(55 citations)**
- Sickafus KE, Minervini L, Grimes RW, et al.; *<u>Radiation</u> tolerance of complex oxides;* <u>Science</u> **289** (2000) 748-751 **(242 citations)**

Ceramic Fuels and Waste Forms Highlights (Modeling Defects and Phase Stability)

- Liu XY, et al., *Mechanism for transient migration of xenon in UO2,* Applied Physics Letters 98 (2011) Article Number: 151902
- Hu SY, et al., *Phase-field modeling of gas bubbles and thermal conductivity evolution in nuclear fuels,* Journal of Nuclear Materials 392 (2009) 292-300
- Liu XY, et al., *Thermodynamics of fission products in dispersion fuel designs - First-principles modeling of defect behavior in bulk and at interfaces,* Nuclear Instruments & Methods In Physics Research, 268 (2010) 3014-3017
- Jiang C, et al., *Predicting from first principles the chemical evolution of crystalline compounds due to radioactive decay: The case of the transformation of CsCl to BaCl,* Physical Review B , 79 (2009) Article Number: 132110

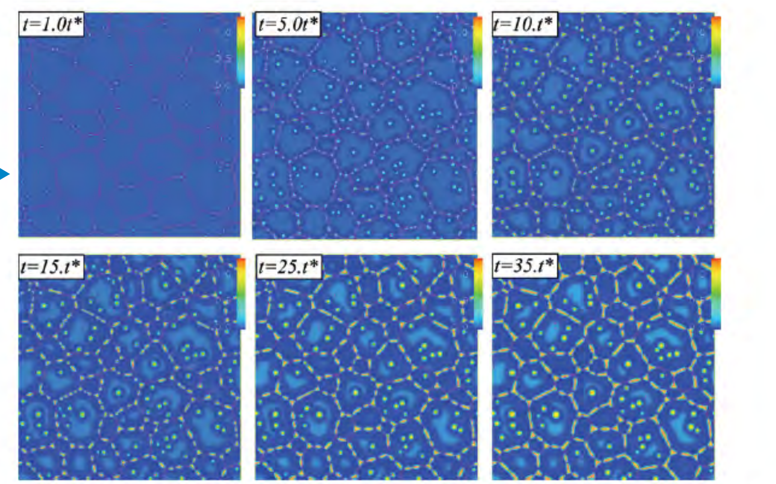
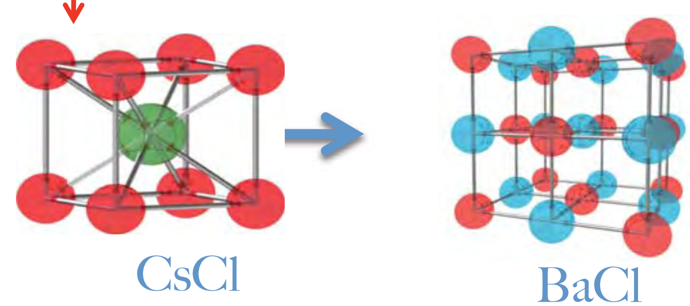
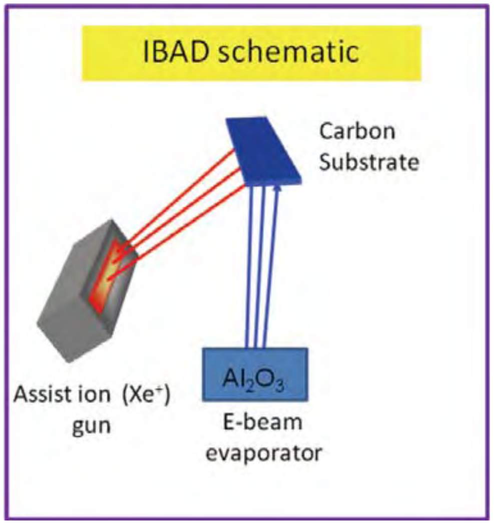


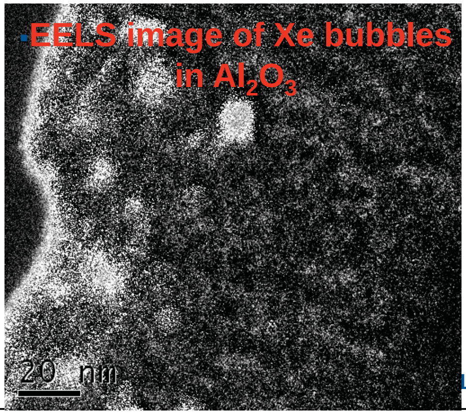
Fig. 6. Time evolution of gas bubbles in a polycrystalline material. The color bar presents the He concentration which varies from 0 to 1.32. The purple lines show the grain boundaries. (For interpretation of the references in colour in this figure legend, the reader is referred to the web version of this article.)



A Novel Technique for Introduction of Fission Gas (Xe) in Solids

- Ion Beam Assisted Deposition (IBAD) concept: simultaneous film growth and low energy ion bombardment
- IBAD concept was verified by using the example of Xe in Al2O3
- IBAD method is highly suitable for fission gas incorporation in nuclear materials
  - Bulk samples with uniform and controllable Xe (and Kr) concentration can be fabricated
  - Any compound (metals, oxides, nitrides, carbides) can be fabricated



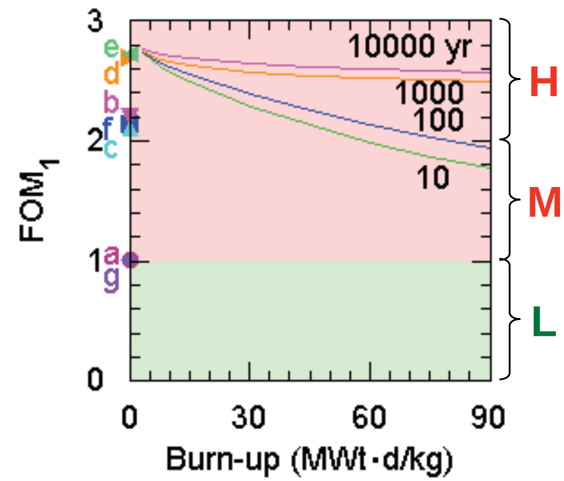


Usov, I et al., Journal of Nuclear Materials 408 (2011) 205-208

Assessment of Material Attractiveness in Advanced Nuclear Fuel Cycles and Waste Forms

- **The Figure Of Merit (FOM) was developed to investigate the validity of various claims regarding fuel cycles and reprocessing schemes being proliferation proof.**
- **The Figure Of Merit (FOM) is a way to characterize material attractiveness in an open forum.**
- **The FOM has been reviewed by LANL and LLNL weapons designers.**

Reactor-Grade Plutonium

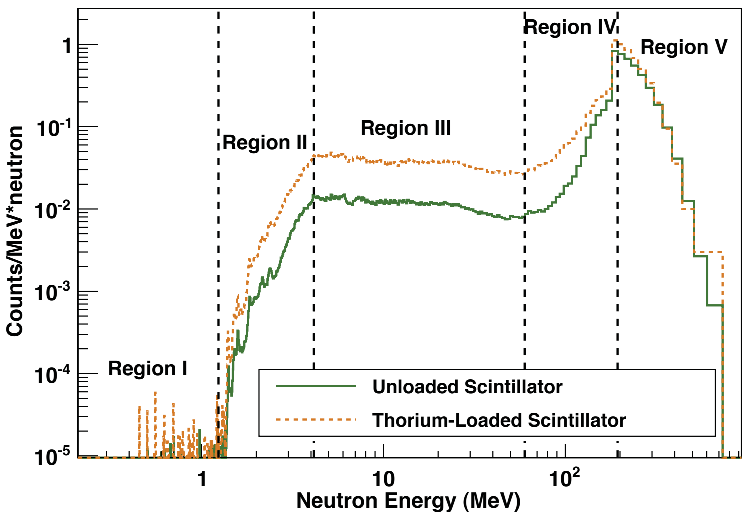


a – LEU (20% ^{235}U)
e – WG-Pu (94% ^{239}Pu)
b – HEU (93% ^{235}U)
f – RG-Pu
c – ^{237}Np
g – ^{238}Pu/^{239}Pu (80:20)
d – ^{233}U (10 ppm ^{232}U)

Bathke (D-5) and Hase (N-4)

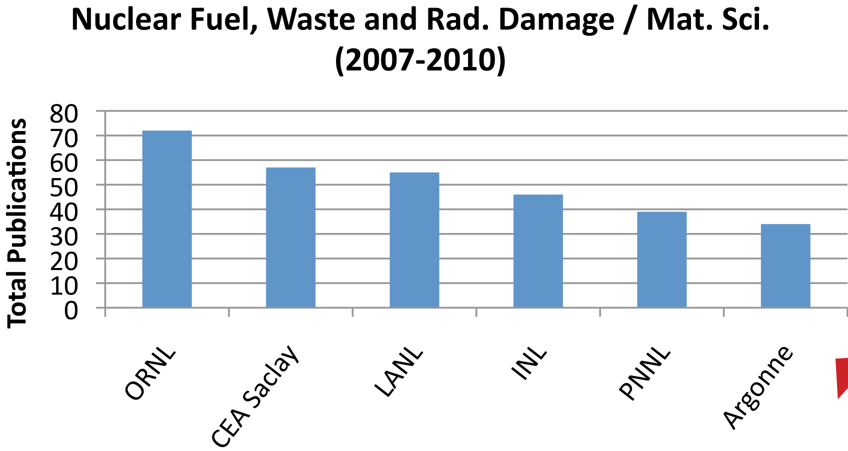
Scintillator Material Development for Detectors Supporting Non-proliferation Activities

- **Goal: an improved neutron flux detector**
  - Rugged and cost-efficient with a high efficiency and a short signal rise time
- **Detector consisted of a liquid scintillating matrix loaded with fissionable ^{232}Th**
- **Thorium-loaded detector showed improved response to neutrons**
  - Higher count rate when exposed to beam line neutrons at LANSCE
  - Delayed decay of fission products complicates attribution of detected events

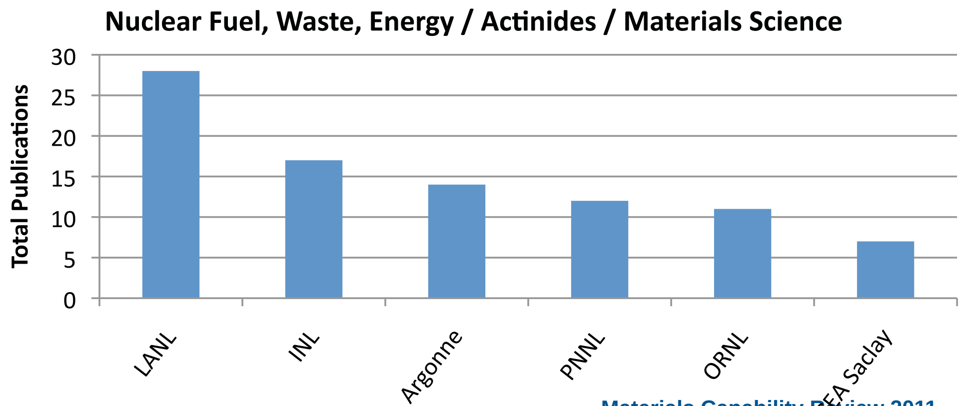


Stange and Esch, N-1

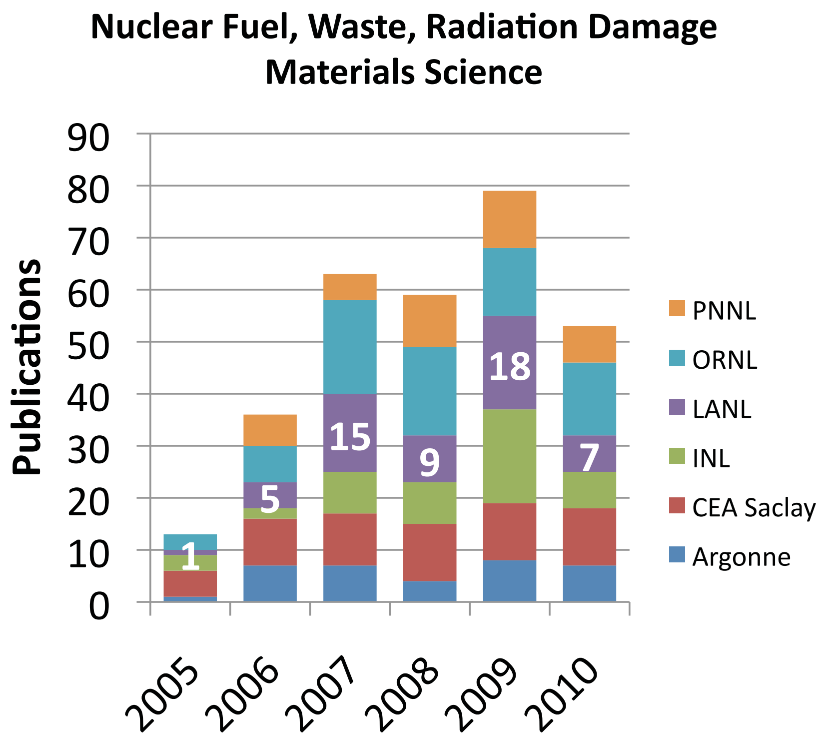
LANL is a strong international leader in publications

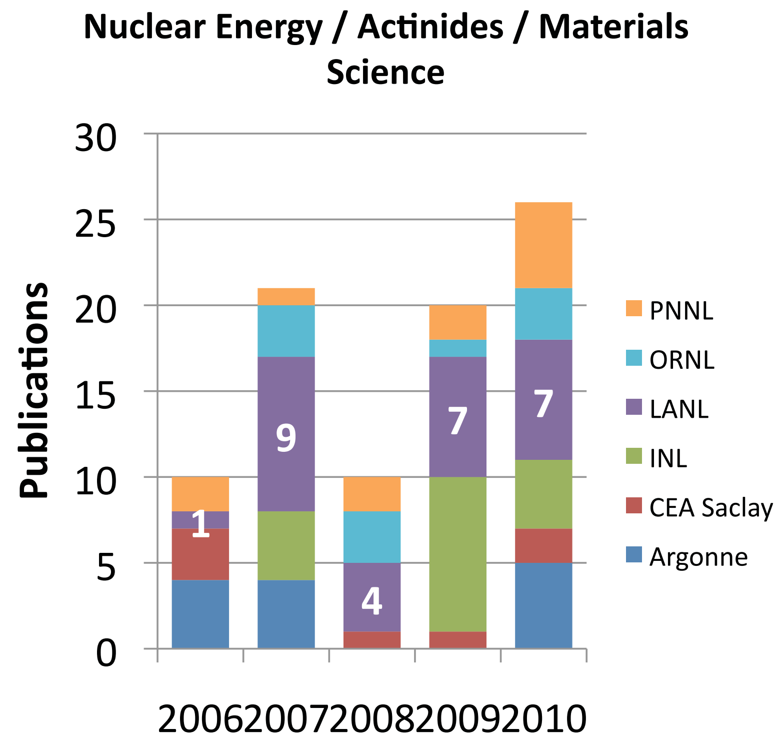


Refine search with actinides



Materials Science Publications in Nuclear Energy Fields are Increasing





Key Science Questions from Materials in Radiation Extremes Deep Dive

- How do we co-design materials for nuclear energy applications?
  - Fuels/wastes to last millennia
  - Radiation tolerant materials
- What are the effects of irradiation on thermophysical properties?
- What are the relevant time and length scales for radiation effects?
- How do interfaces and microstructure control irradiation response of materials? And how can we control them?
- What are the appropriate in-situ measurement capabilities for measuring damage evolution from irradiation cascades to microstructural defects?
- What computational capability is required to model these processes over the range of length and time scales?
- How do accelerated aging and non-accelerated aging experiments correlate?
- What are the separations challenges that we need to “improve” to diminish nonproliferation concerns, improve economics and improve safety?

Strategic Implementation Opportunities

- **Two year timeframe:**
  - Explore existing irradiation capabilities at LANL (IBML, IPF, DAHRT, WNR) and elsewhere (SNL, Japan, ATR, etc.)
  - Establish more robust partnership between radioactive materials-centric facilities – MSL, Sigma, CMR, TA55, TA35 and LANSCE and leverage these facilities for new programs in materials.
- **Five year timeframe:**
  - Establish capability to translate from fundamental science to technology on a 5 year time frame
  - Better integration of experimental and IS&T capabilities in materials design, enabling co-design
  - Ability to design, synthesize and predict long-term behavior of simple 2 component materials (radiation tolerant and fuels).
- **Ten year timeframe:**
  - Ability to accurately model long-term aging behavior based on accelerated aging experiments
  - Ability to design, synthesize and predict long-term behavior of complex, engineering materials (radiation tolerant and fuels).

Presentations and Posters

- **Presentations:**
  - Chris Stanek – “Lower Length Scale Simulations of Fission Gas and Thermal Transport in Oxide Nuclear Fuel”
  - Andy Nelson – “Development of Experimental Techniques to Augment Theory and Simulation of Thermal Transport in Oxide Nuclear Fuel”

Poster Presentations

- **Metal Fuels**
  - Dave Dombrowski – "LEU U-10Mo Research Reactor Fuel Development and Scale Up"
  - Dave Korzekwa – "Process Modeling of Plutonium and Uranium Casting"
  - Don Brown – "Residual Stresses in Aluminum Clad Uranium-10wt%Molybdenum Fuel Plates"
  - Sven Vogel – "Chemical Segregation of U-10Mo Fuel Foils during Simulated Bonding Cycles Using Neutron Diffraction"
- **Ceramic Fuels and Waste Forms**
  - Jeremy Mitchell – "Sintering of Mixed-Oxide Fuel Pellets"
  - Boris Dorado – "Radioparagenesis: Robust Nuclear Waste Form Design and Novel Material Discovery"
  - Eric Bauer – "Plutonium Science and Research Strategy: Use of Special Isotopes – ^{242}Pu"
- **Non-proliferation**
  - Esch – "Fissionable Scintillators for Measuring Neutron Flux"
  - Bathke – "Material Attractiveness in Nuclear Fuel Cycles"

LANSCE relevance

Lower length scale simulations of fission gas and thermal transport in oxide nuclear fuel

C.R. Stanek (MST-8)

As is well known, the performance of nuclear fuel is strongly related to the transport behavior of fission gases, such as xenon. If fission gases are retained, the fuel swells and may mechanically interact with the clad. Whereas, if the fission gases are released the plenum pressure increases. Both situations increase the probability of clad failure and therefore must be understood for fuel qualification. The complexity of fission gas behavior has to date prevented a systematic understanding. In this talk, we present recent simulations of Xe in UO_2 that contribute to improved understanding of fission gas behavior. Specifically, we have employed a number of different “lower length scale” simulation techniques and built upon seminal work from the Harwell Theoretical Physics group to consider bulk diffusion of Xe in UO_2 as a function of nonstoichiometry using density functional theory, as well as pair potentials to consider the effects of grain boundary structure of gas transport. By considering the relationship between 1, 2, and 3 dimensional defects (i.e. dislocations, grain boundaries and bubbles) with zero dimensional defects (i.e. fission gas atoms), we are able to simulate fission gas transport as a function of idealized microstruture. Such insight can subsequently be used to improve meso- and engineering-scale models, examples of which will also be discussed. Additionally, the extension of this approach will be demonstrated by several examples of oxide fuels with variable compositions. Finally, we employ a similar approach to improve understanding of oxide fuel thermal conductivity. Since many materials properties that govern fuel performance are strongly temperature dependent, understanding how the thermal conductivity evolves during the lifetime of the fuel is important for fuel performance simulations. Preliminary results of the dependence of grain boundary structure on Kaptiza resistance will be shown.

Lower Length Scale Simulations of Fission Gas and Thermal Transport in Oxide Nuclear Fuel

C.R. Stanek

MST-8, Structure and Property Relations

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Simon Phillpot

Pedro Peralta

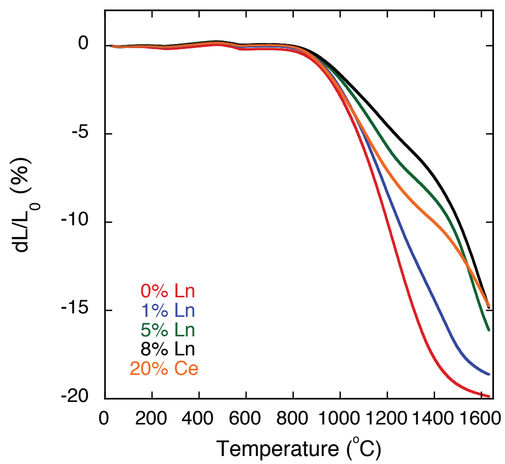
Ben Hanken
Mark Asta
Alex Navrotsky

Outline

- **Motivation/Approach –** Improve understanding of the interaction between multidimensional defects by combining modeling (including contributions to engineering scale codes) with experiment.
- **Results**
  - **"Natural Defects":** UO_2 nonstoichiometry, grain boundaries, Ln impurities
  - **"Radiation-Produced Defects":**
    - *Fission gas release* (first principles study of bulk Xe diffusion compared to exp. data; atomistic study of fission gas interaction with microstructure; "upscaling" both the above to meso- and engineering scale)
    - *Thermal conductivity* (atomistics: implementation of long range forces for consideration of microstructure; Upscaling of atomistics to meso- and engineering scale)

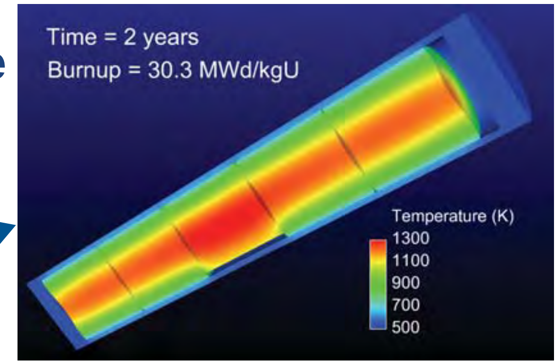
Simplified Approach:

Strategic Problem Definition Enables Integration and Improves Relevance



Experiments (FCRD, EFRC)

Fuel Performance Code (CASL, NEAMS)



microstructure

What?: Informed simulation of effect of microstructure on fuel performance and synthesis

How?: mesoscale simulation techniques

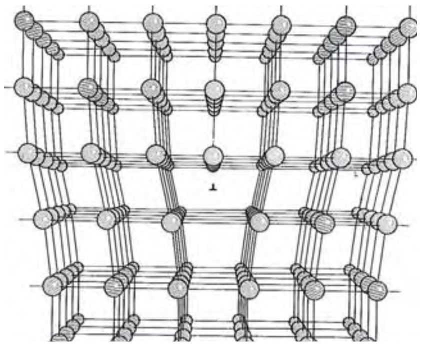
Single crystal

Idealized polycrystal

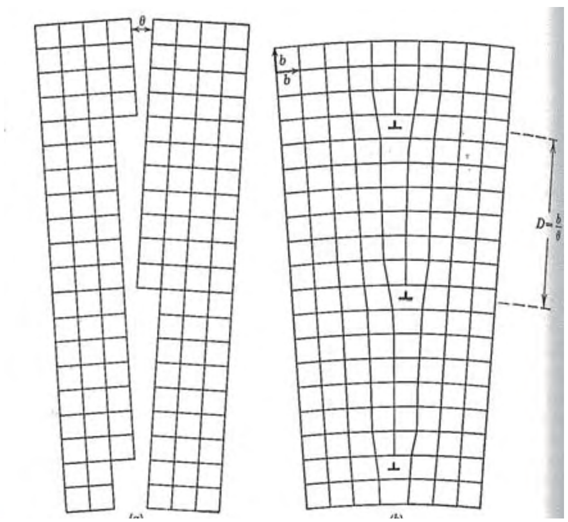
What?: Thermal and mass transport as a function of composition, nonstoichiometry, GB structure, dislocation structure, etc.

How?: Atomistics (MD, DFT, etc), thermochemical models, etc.

Recurring Theme = Interaction Between Multidimensional Defects



From C.A. Wert and R.M. Thomson "Physics of Solids" (1964).



From W.T. Read Jr. and W. Shockley, in F. Seitz "Imperfections in Nearly Perfect Crystals" (1952).

Understanding the interaction between defects may help reveal how materials properties evolve with irradiation.

1. Natural defects:
   (a) Grain boundaries.
   (b) Dislocation lines.
   (c) Closed pores in the as-fabricated fuel.
   (d) Impurities in the solid.
2. Radiation-produced defects:
   (a) Vacancy clusters (perhaps stabilized by a few gas atoms). A fission track is especially rich in this type of trap.
   (b) Dislocation loops formed by condensation of excess interstitial atoms.
   (c) Fission-gas bubble.
   (d) Solid fission-product precipitates (e.g., the noble metal and alkaline earth oxide phases).

D.R. Olander, "Fundamental Aspects of Nuclear Reactor Fuel Elements" (1976)

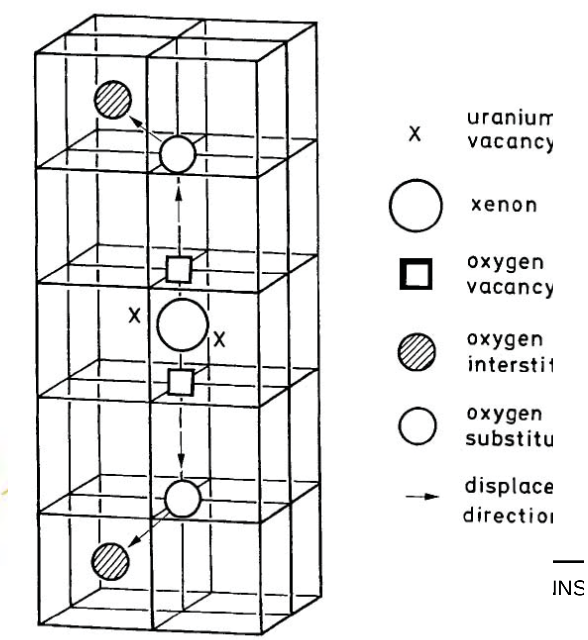
Why Now?

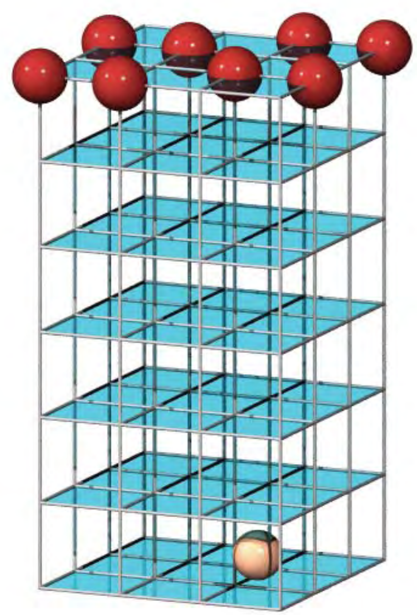
Olander elucidated many of the important issues over 30 years ago.

Original simulation study: Catlow, “Fission Gas Diffusion in Uranium Dioxide,” Proc. Roy. Soc. London A **364** (1978) 473.

Xe diffusion mechanism proposed by Ball and Grimes (J. Chem. Faraday Trans. **86(8)** (1990) 1257) from **pair potentials**.

Surface segregation: C.R. Stanek, et al., J. Phys.: Cond. Matter **16** (2004) S2699.





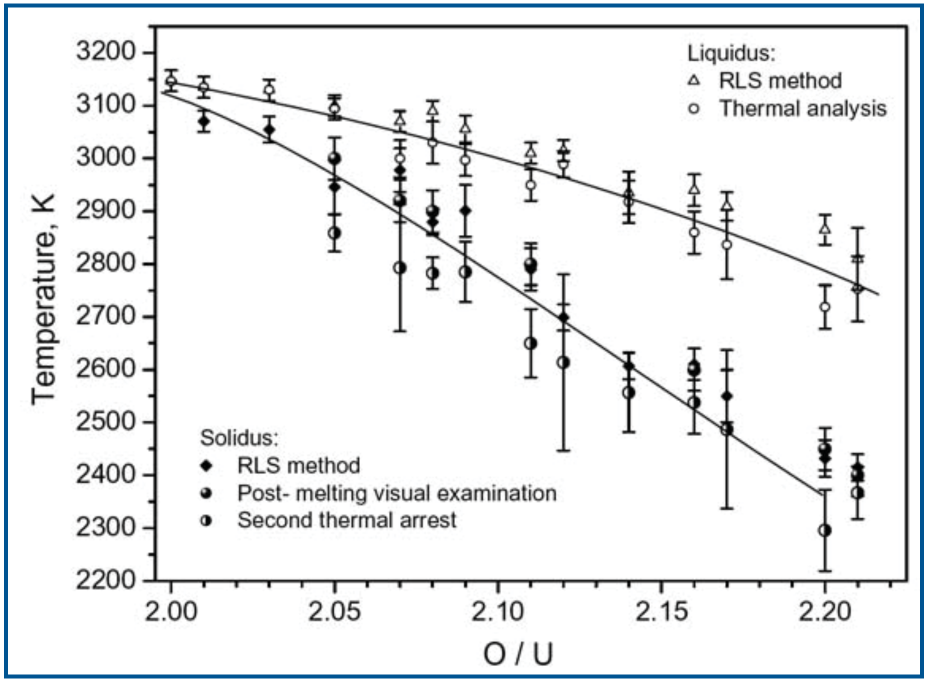
Two important, recent advances – both benefit from high performance computing:

DFT for improved quantification

Pair potentials for *relevant* microstructural features

Results Part 1: Natural Defects

UO_2 Nonstoichiometry



D. Manara, et al. *J. Nucl. Mater.* **342** (2005) 148.

D. A. Andersson, T. Watanabe, C. Deo, and B. P. Uberuaga, "Role of di-interstitial clusters in oxygen transport in UO_{2+x} from first principles," *Phys. Rev. B* **80**, 060101(R) (2009).

D. A. Andersson, J. Lezama, B. P. Uberuaga, C. Deo, and S. D. Conradson, "Cooperativity among defect sites in AO_{2+x} and A_4O_9 (A=U,Np,Pu): Density functional calculations", *Phys. Rev. B* **79**, 024110 (2009).

D. A. Andersson, F. J. Espinosa-Faller, B. P. Uberuaga and S. D. Conradson, "Configurational stability and migration of large oxygen clusters in UO_{2+x}: Density functional theory calculations", to be submitted.

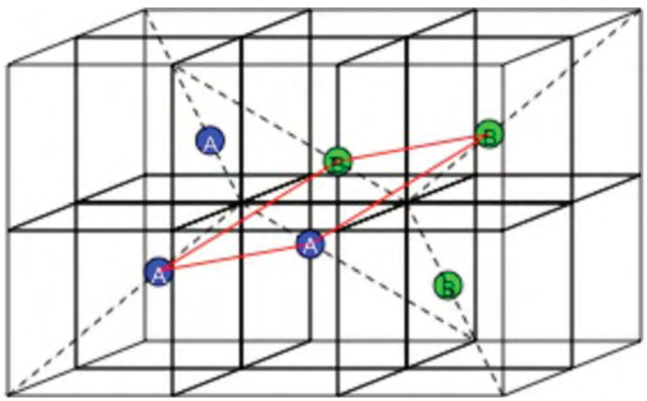
Results Part 1: Natural Defects

UO_2 Nonstoichiometry

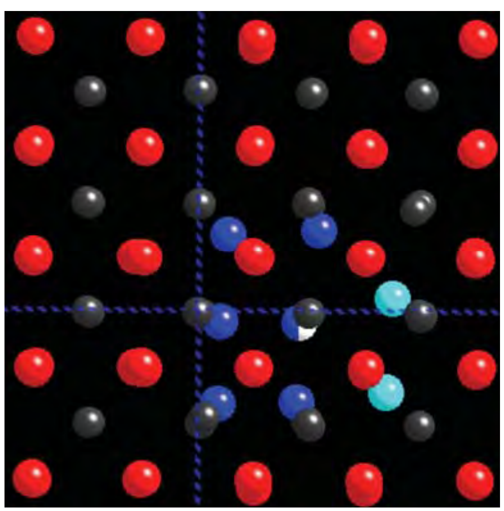
The split quad-interstitial (I_4^X) is the most stable state of excess oxygen ions in UO_{2+x} - more stable than the cuboctahedrons proposed from experiments.

Split quad-interstitial (I_4^X)

Two split di-interstitials (blue and green) make up a stable cluster in AnO_{2+x}.

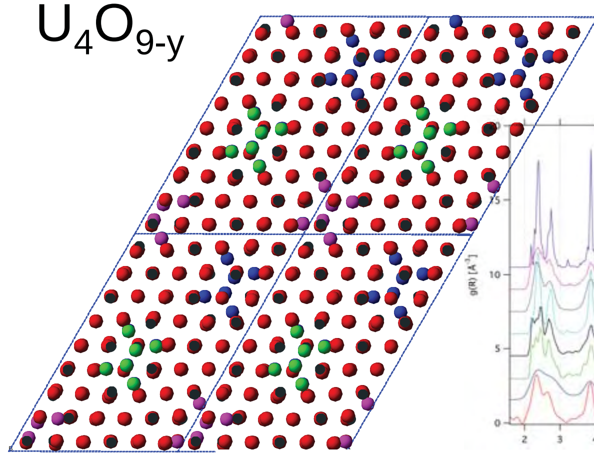


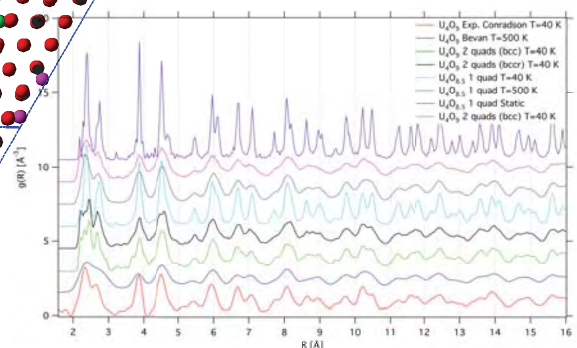
Enables new diffusion mechanism



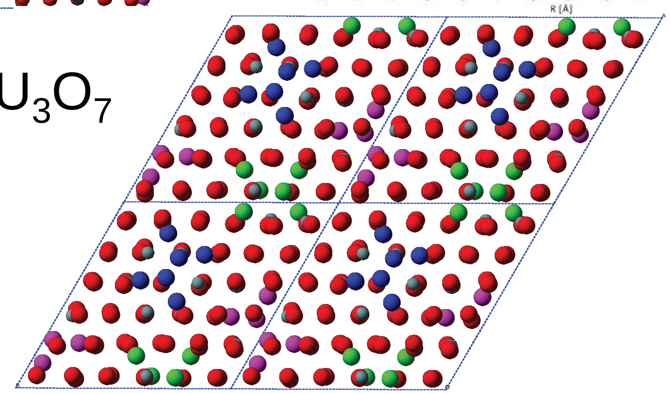
New models for U_4O_{9-y} and U_3O_7 consistent with DFT being developed by combining theory and neutron diffraction

U_4O_{9-y}





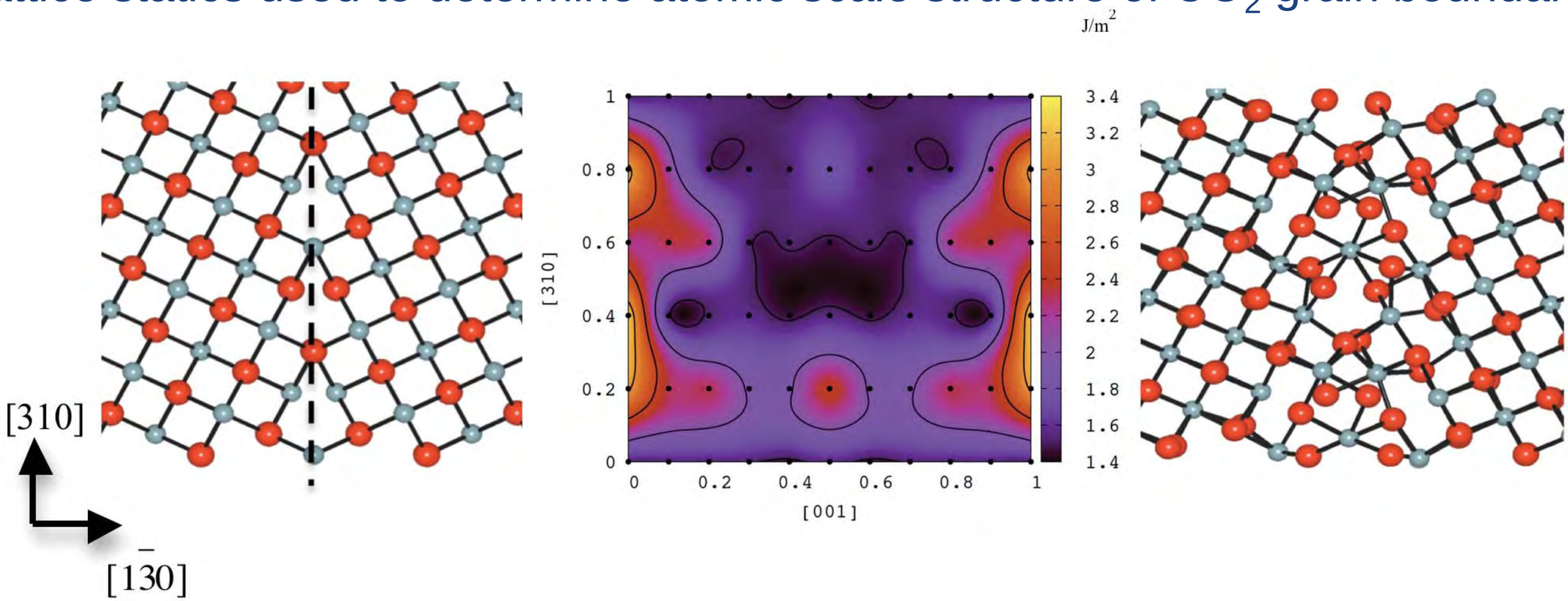
U_3O_7



Results Part 1: Natural Defects

UO_2 Grain Boundary Structure

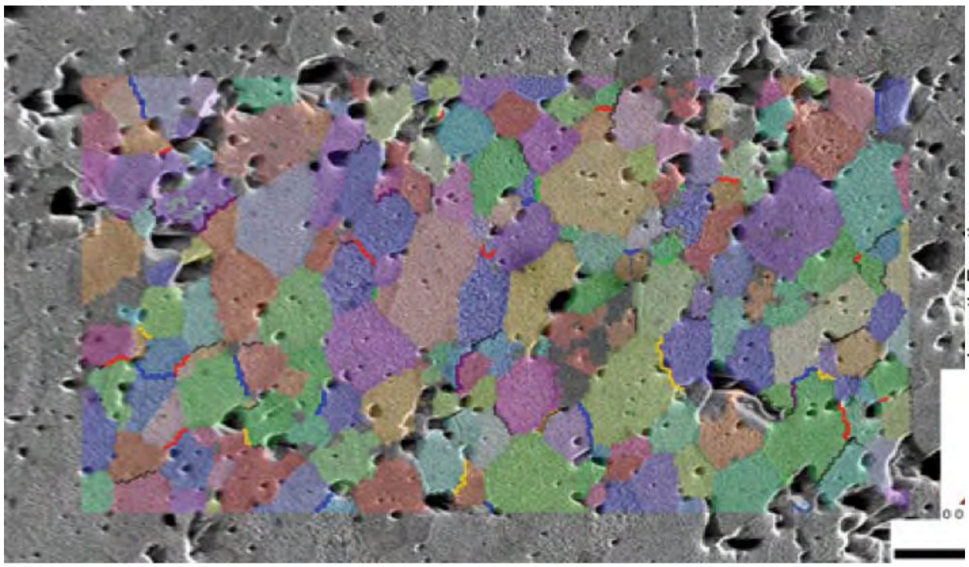
Lattice statics used to determine atomic scale structure of UO_2 grain boundaries

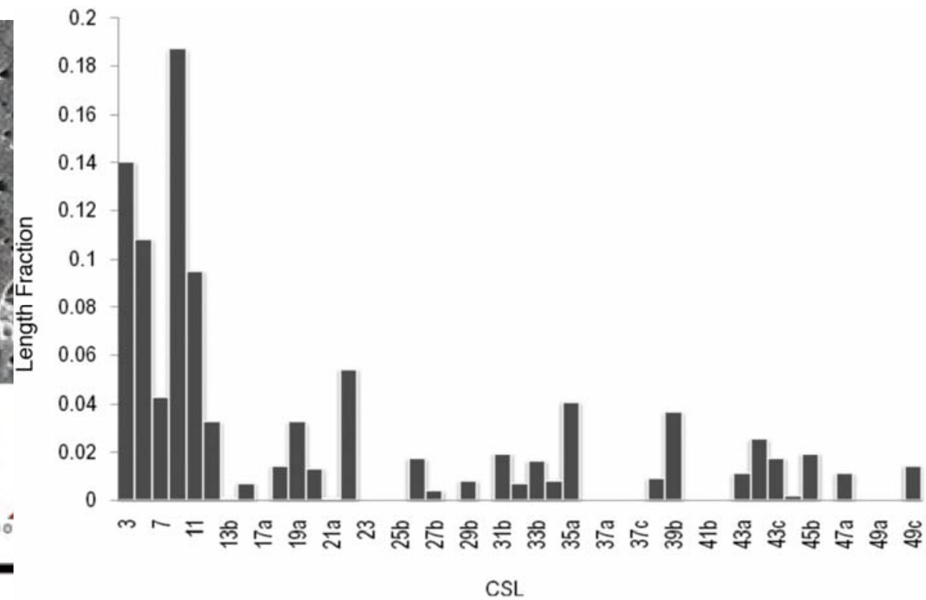


P.V. Nerikar, K. Rudman, T. G. Desai, D. Byler, C. Unal, K. J. McClellan, S. R. Phillpot, S. B. Sinnott, P. Peralta, B. P. Uberuaga, and C.R. Stanek, “Grain Boundaries in UO_2: Scanning Electron Microscopy Experiments and Atomistic Simulations,” in press *J. Amer. Ceram. Soc.* (2011).

Results Part 1: Natural Defects
UO_2 Grain Boundary Structure

Complementary characterization experiments on FCRD pellets (pre-ATR irradiation) focus atomistics





P.V. Nerikar, K. Rudman, T. G. Desai, D. Byler, C. Unal, K. J. McClellan, S. R. Phillpot, S. B. Sinnott, P. Peralta, B. P. Uberuaga, and C.R. Stanek, "Grain Boundaries in UO_2: Scanning Electron Microscopy Experiments and Atomistic Simulations," in press *J. Amer. Ceram. Soc.* (2011).

Results Part 1.5: Natural-ish Defects

Ln^{3+} Solution in UO_2

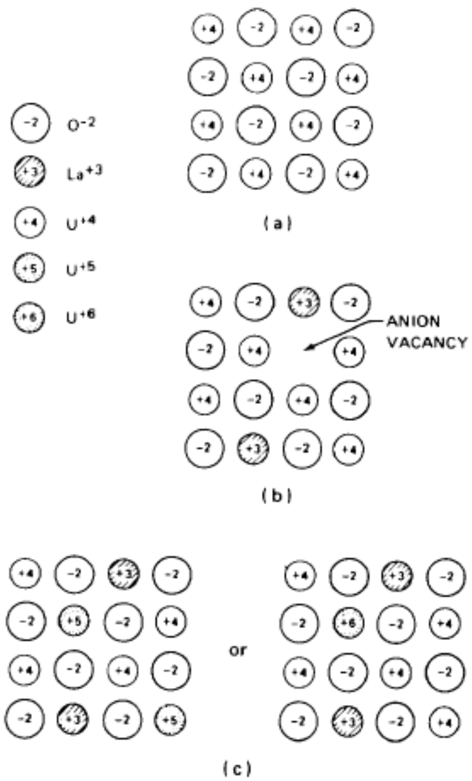
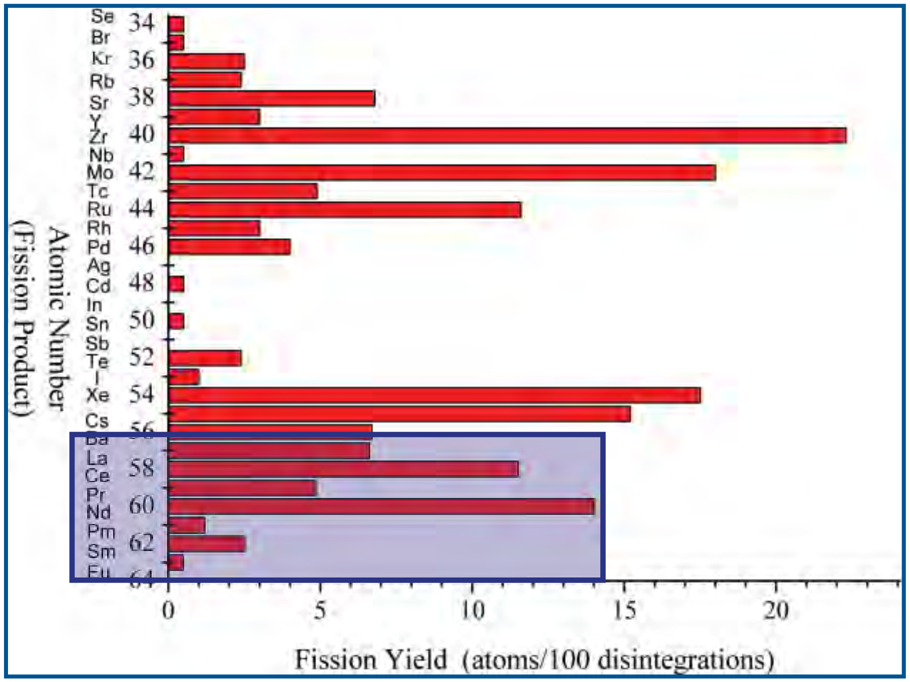


Fig. 12.8 Two-dimensional representation of methods of maintaining electrical neutrality when La^{3+} is introduced into UO_2. (a) Perfect UO_2 lattice. (b) Removal of oxygen ions. (c) Oxidation of U^{4+} to U^{5+} or U^{6+}.

The different solution mechanism possibilities are well known – but determination difficult.

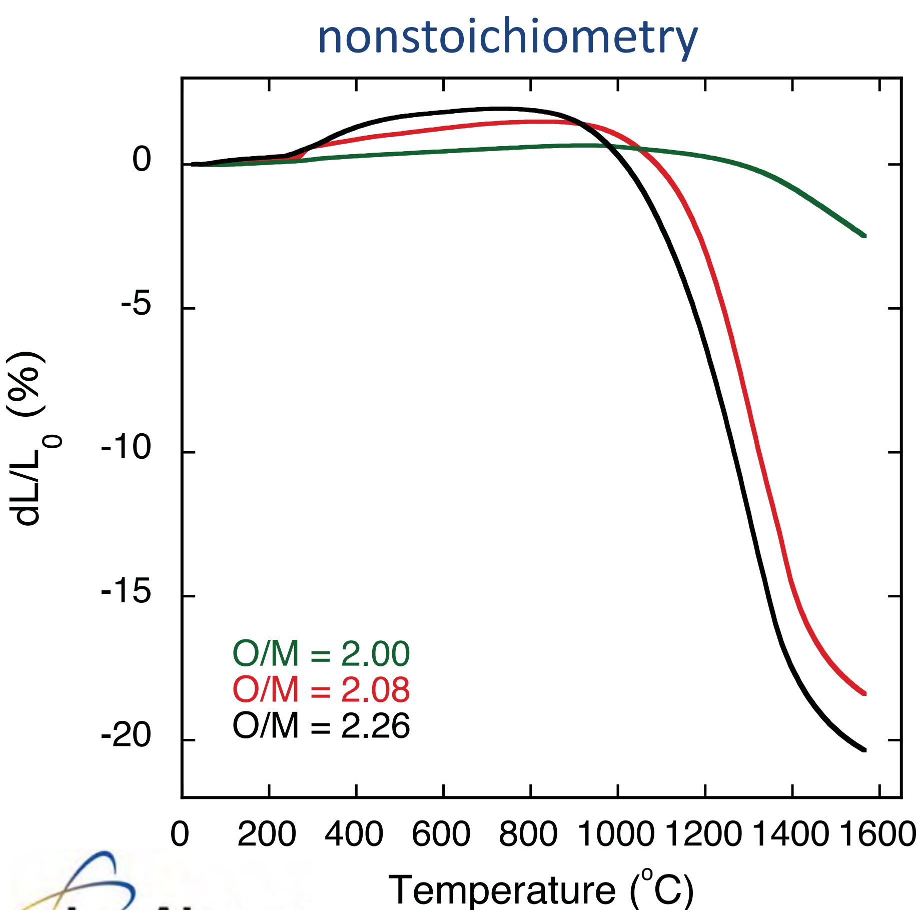
Implication is increased uncertainty of nonstoichiometry

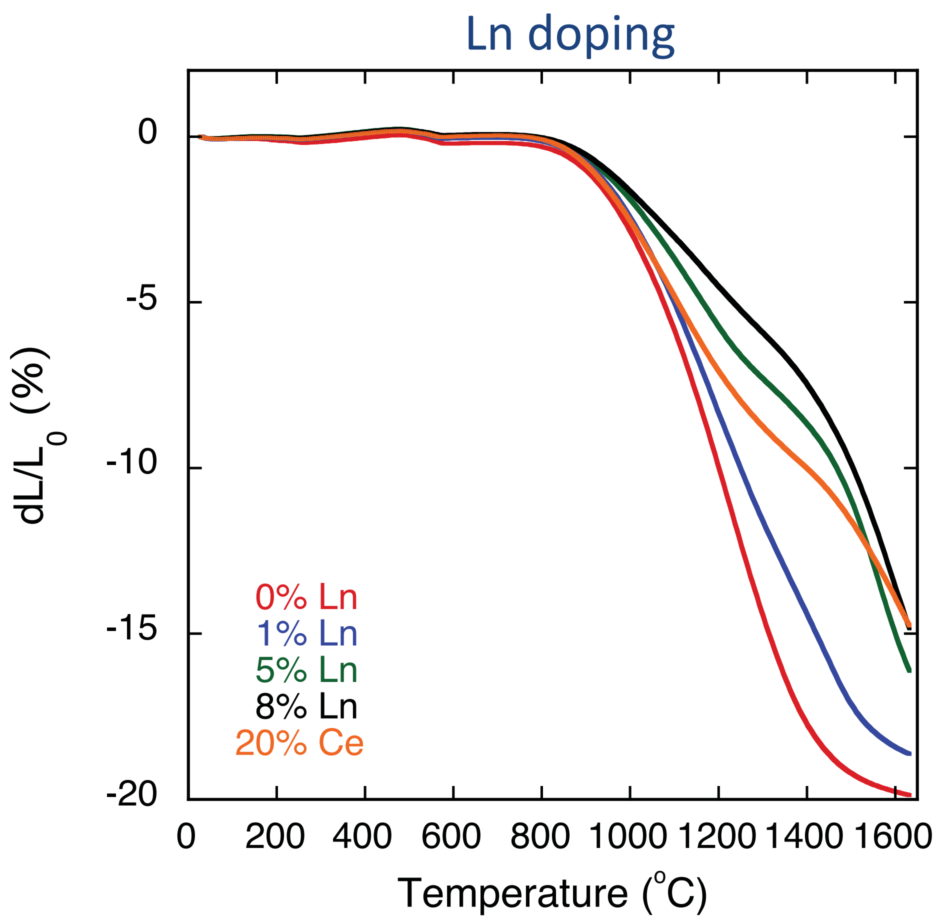


Results Part 1.5: Natural-ish Defects

Ln^{3+} Solution in UO_2

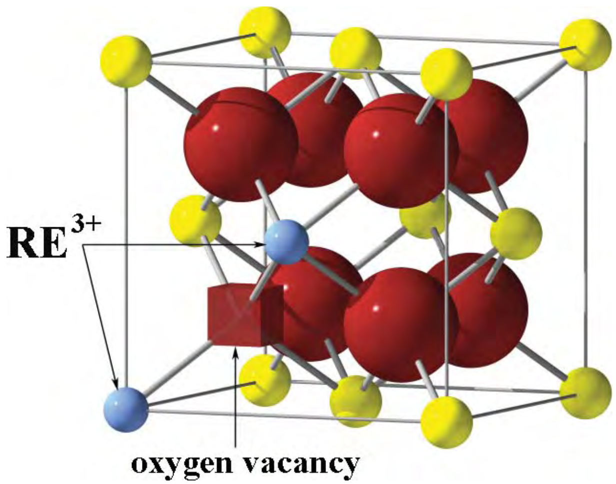
Similarity between sintering curves for nonstoichiometric and Ln-doped UO_2

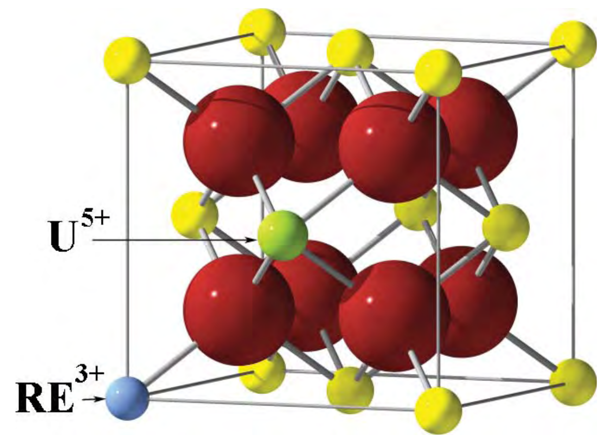




Results Part 1.5: Natural-ish Defects

Ln^{3+} Solution in UO_2





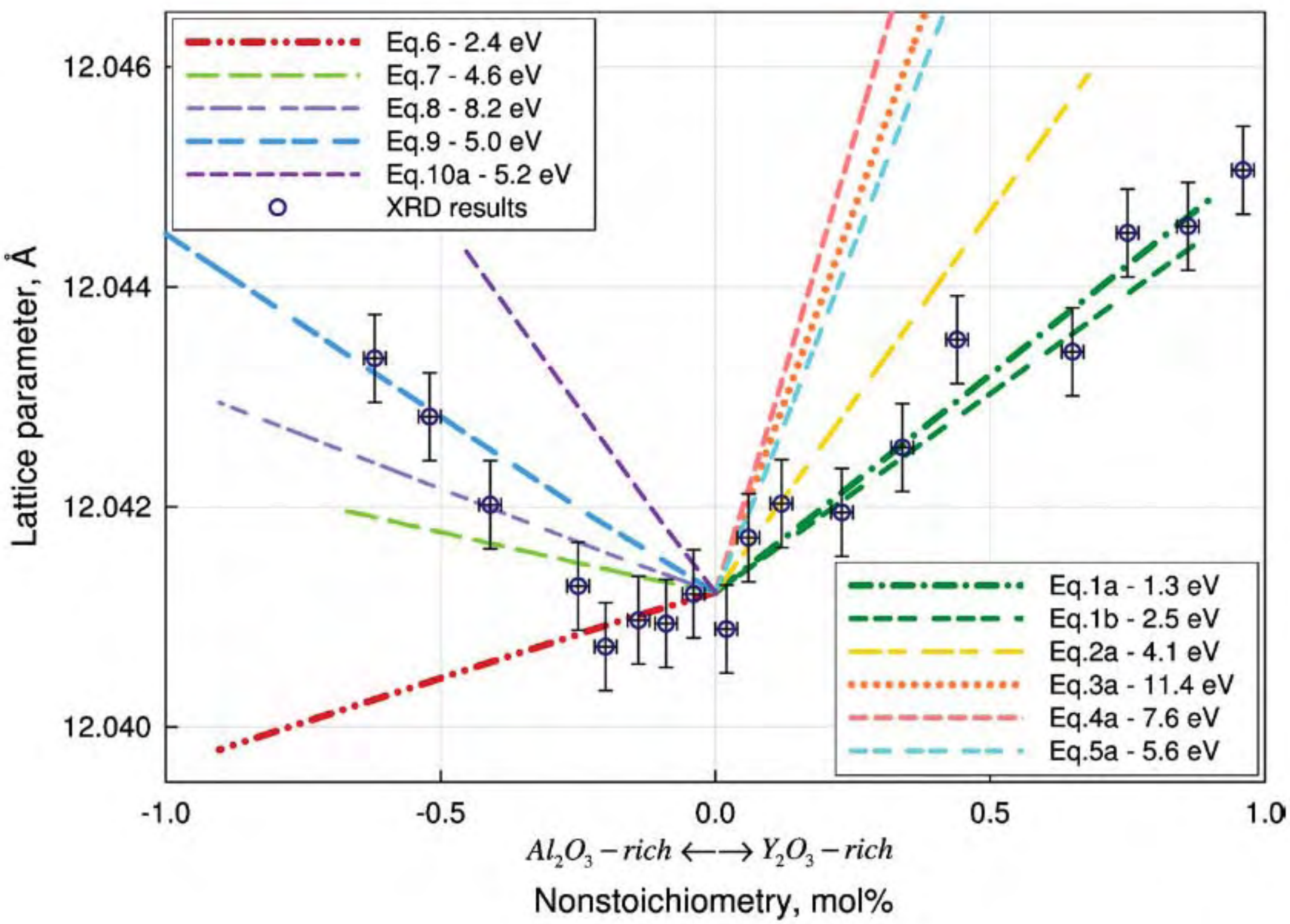
$$2U_U^X + O_O^X + RE_2O_3 \rightleftharpoons 2RE'_U + V_O^{\bullet\bullet} + 2UO_2$$

$$8U_U^X + 2RE_2O_3 \rightleftharpoons 4RE'_U + 4U_U^{5+\bullet} + 3UO_{2+x}$$

Although this study considers only Ln^{3+}, the results and implication of are likely applicable to minor actinides such as Am.

Results Part 1.5: Natural-ish Defects

Ln^{3+} Solution in UO_2



Defect volumes provide a alternative mechanism for comparison.

For example, by comparing atomistic defect volumes to experimental lattice parameter variation - we have determined non-stoichiometry mechanisms for YAG.

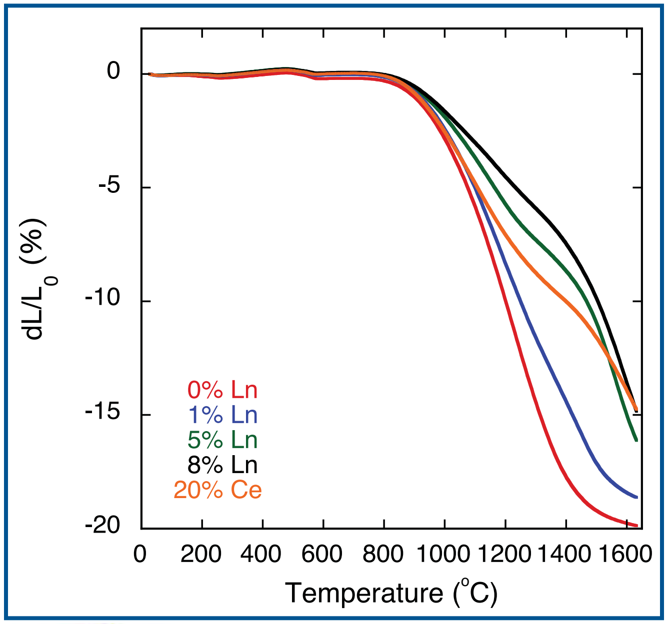
A.R. Patel *et al., Appl. Phys. Lett.* **93** (2008) 1911902

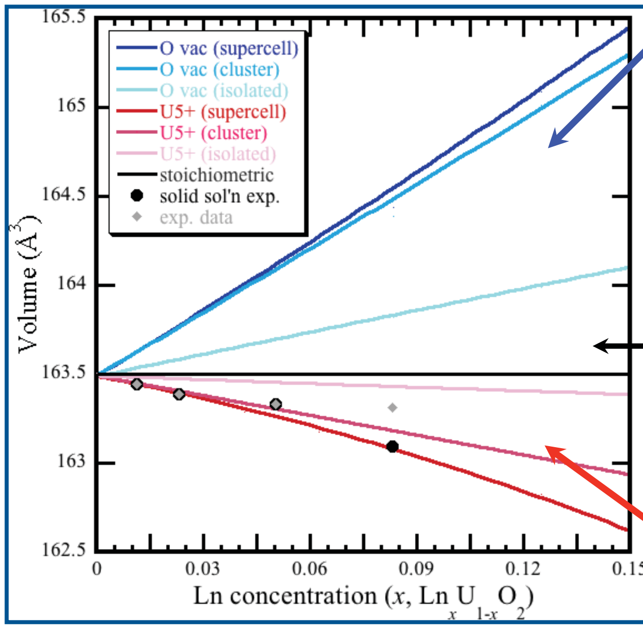
Results Part 1.5: Natural-ish Defects

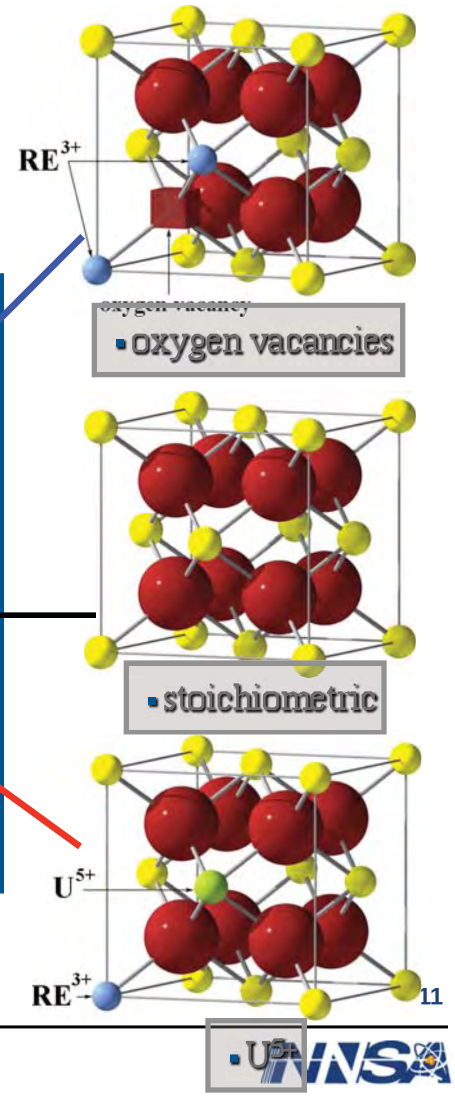
Ln^{3+} Solution in UO_2

Known from Olander that La additions to UO_2 result in a lattice expansion, while Y results in contraction

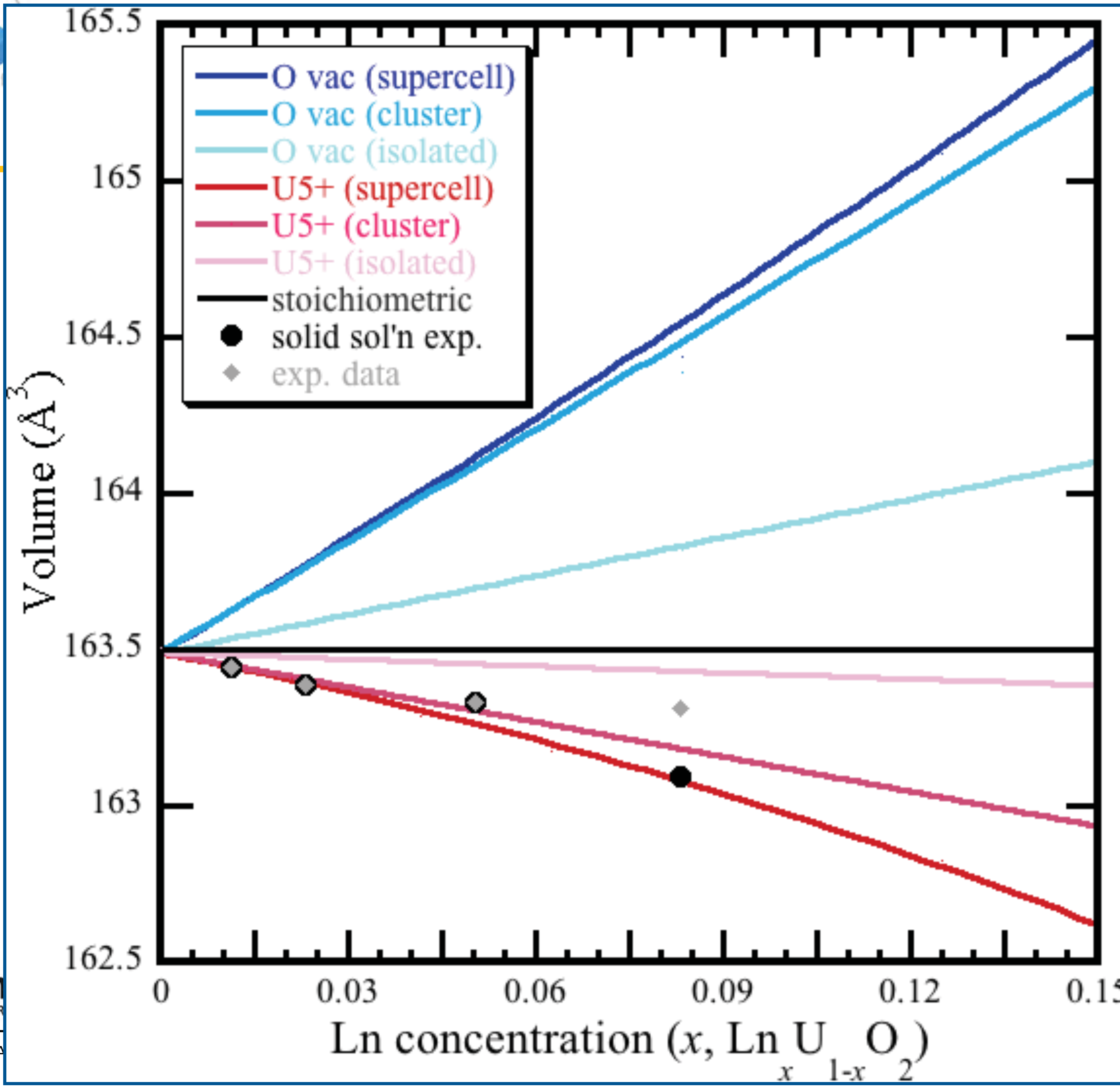
Effect of 6% La, 16% Pr, 25% Ce and 53% Nd?





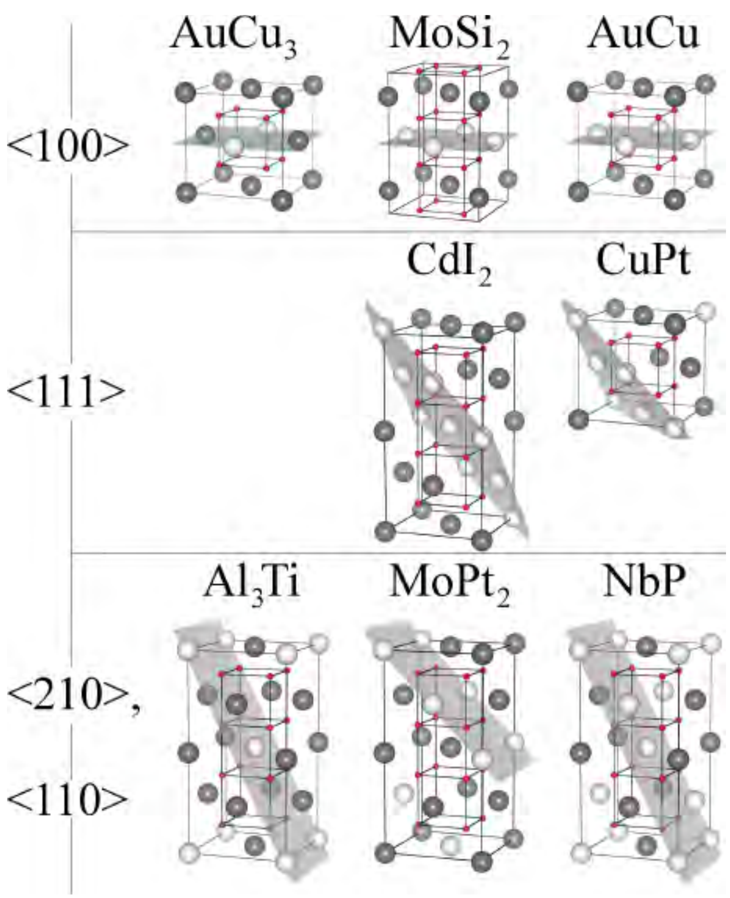


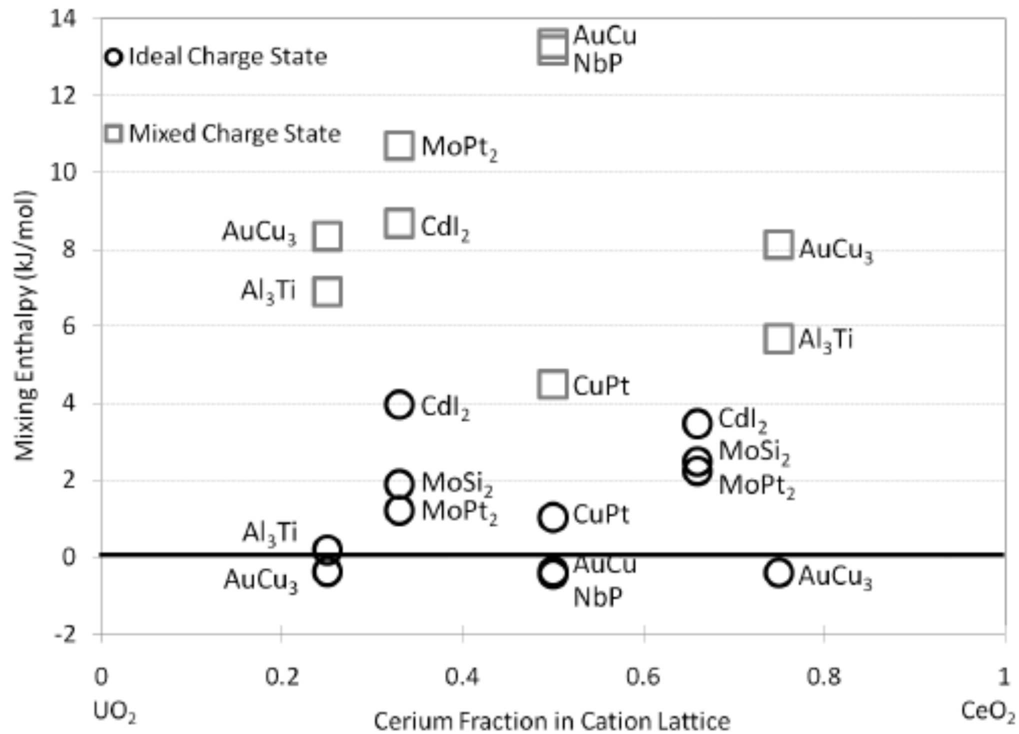
Los Alamos


165.5
165
164.5
164
163.5
163
162.5
0
0.03
0.06
0.09
0.12
0.15
Volume (Å^3)
Ln concentration (x, Ln_x U_1-x O_2)
O vac (supercell)
O vac (cluster)
O vac (isolated)
U5+ (supercell)
U5+ (cluster)
U5+ (isolated)
stoichiometric
solid sol'n exp.
exp. data

Results Part 1.5: Natural-ish Defects

$Ce^{3+/4+}$ Solution in UO_2





B.E. Hanken, C.R. Stanek, N. Grønbech-Jensen and M. Asta, “Computational study of the energetics of charge and cation mixing in $U_{1-x}Ce_xO_2$,” in review *Physical Review B*.

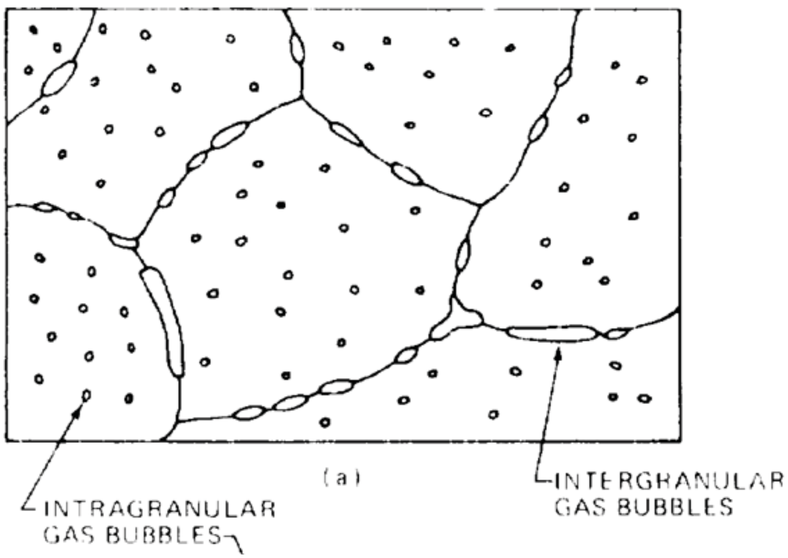
Results Part 2: Radiation Produced Defects
Fission Gas Release Models in UO_2

Simplest model for fission gas release is the **Booth model**.

Relies on an "Equivalent Sphere" – where bulk diffusion is rate limiting since grain boundaries serve as an infinite sink, and all gas is released from boundary.

Booth model does not account for trapping or re-solution.

Forsberg-Massih model builds upon Booth sphere to include trapping/ resolution.

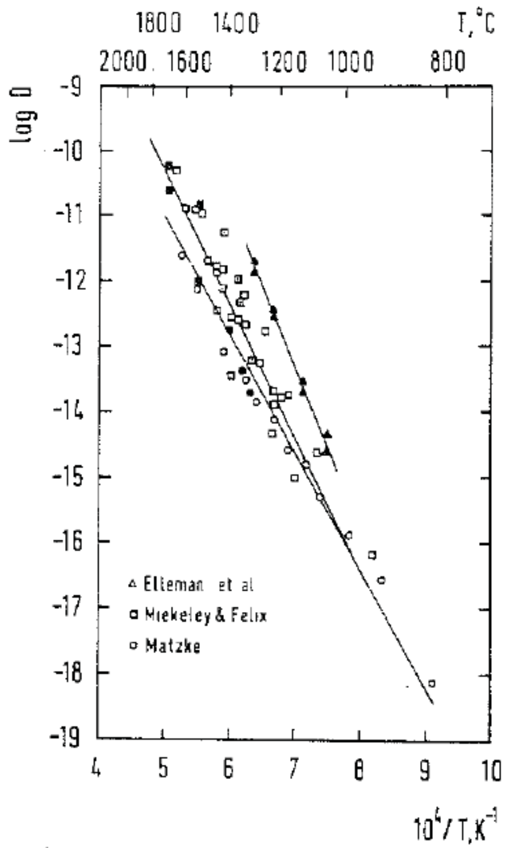


D.R. Olander, "Fundamental Aspects of Nuclear Reactor Fuel Elements" (1976)

Results Part 2: Radiation Produced Defects

DFT Calculations of Bulk Xe Diffusion in UO_2

Experiments, e.g. Hj. Matzke, *Radiation Effects* **53** (1980) 219.



ΔH_A Xe:

ΔH_A=6.0 eV UO_{2-x}.

ΔH_A=3.9 eV UO_2.

ΔH_A=1.7 eV UO_{2+x}.

ΔH_A U:

ΔH_A=7.8 eV UO_{2-x}.

ΔH_A=5.6 eV UO_2.

ΔH_A=2.6 eV UO_{2+x}.

Easier to form U vacancy for x>0 and x=0 than for x<0.

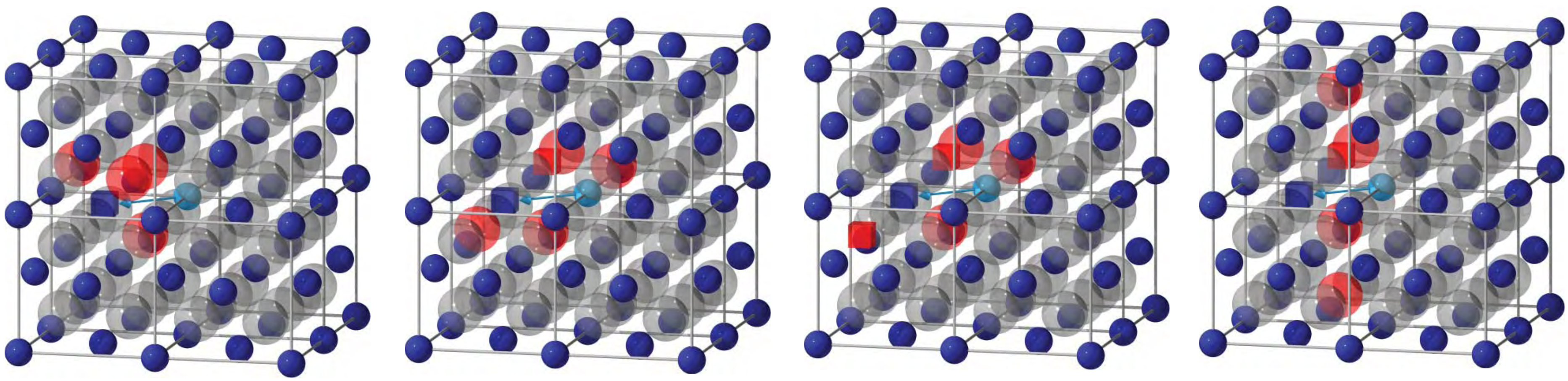
- Effective activation energies determined decades ago, but mechanistic aspects not understood.

- Not of academic interest, but required for formulating accurate and predictive diffusion models that account for irradiation.

$$D = D_0 \exp\left(-\frac{\Delta H_A}{k_B T}\right)$$

Results Part 2: Radiation Produced Defects
DFT Calculations of Bulk Xe Diffusion in UO_2

Diffusion of U vacancies is the rate limiting step for Xe transport. Barriers for U vacancy diffusion ranges from 2 to 5 eV depending on cluster configuration.



"Xe and U transport in $UO_{2\pm x}$: Density functional theory calculations," D.A. Andersson, B.P. Uberuaga, P.V. Nerikar, C. Unal and C.R. Stanek – in review *Physical Review B*.

Results Part 2: Radiation Produced Defects

DFT Calculations of Bulk Xe Diffusion in UO_2

	Calc.	Exp. [1]	Fitted
E_F	3.44	3.0-4.0	3.26
E_S	5.70	6-7	5.26
$E_B(Xe_{UO2})$	2.90	--	3.03
$E_B(Xe_{UO})$	1.60	--	1.87
$E_B(Xe_U)$	1.22	--	0.81
$E_M(V_U)$	3.77	2.4*	--
$E_M(V_{UO2})$	3.94	--	--
$E_M(V_{U2})$	2.18	--	--
$E_M(V_{U2O})$	2.67	--	--

	Calc.	Exp [1]
$E_a^U(UO_{2-x})$	8.15	7.8
$E_a^U(UO_2)$	6.03	5.6
$E_a^U(UO_{2+x})$	2.58	2.6
$E_a^{Xe}(UO_{2-x})$	6.57	6.0
$E_a^{Xe}(UO_2)$	4.42	3.9
$E_a^U(UO_{2+x})$	1.36	1.7

[3] Hj. Matzke, in Diffusion Processes in Nuclear Materials, ed. R. P. Agarwala, North-Holland, Amsterdam, 1992.

* Incorrect assignment.

Results Part 2: Radiation Produced Defects

DFT Calculations of Bulk Xe Diffusion in UO_2

Interstitial diffusion mechanism relevant if thermal or radiation-induced defects (i.e. vacancies) are present only in low concentrations.

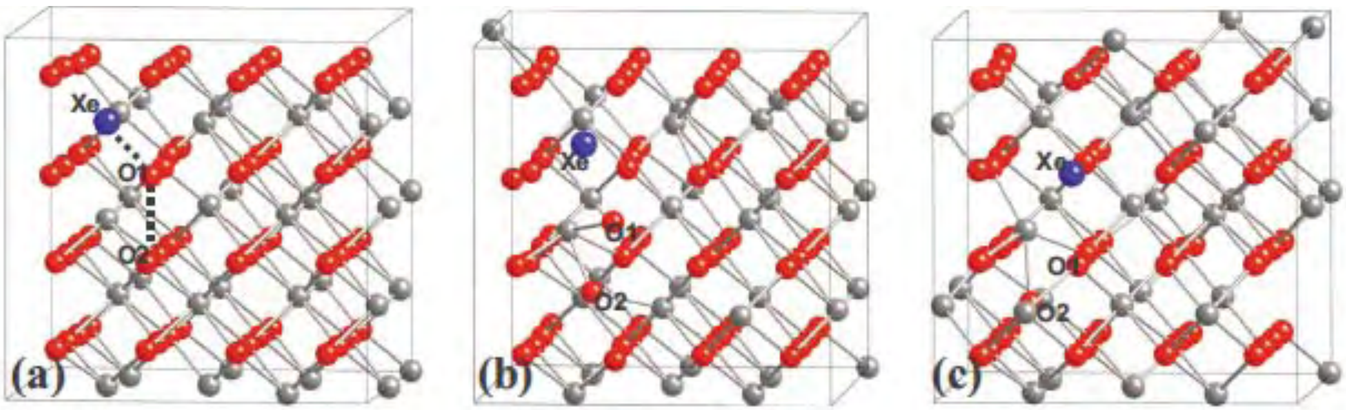
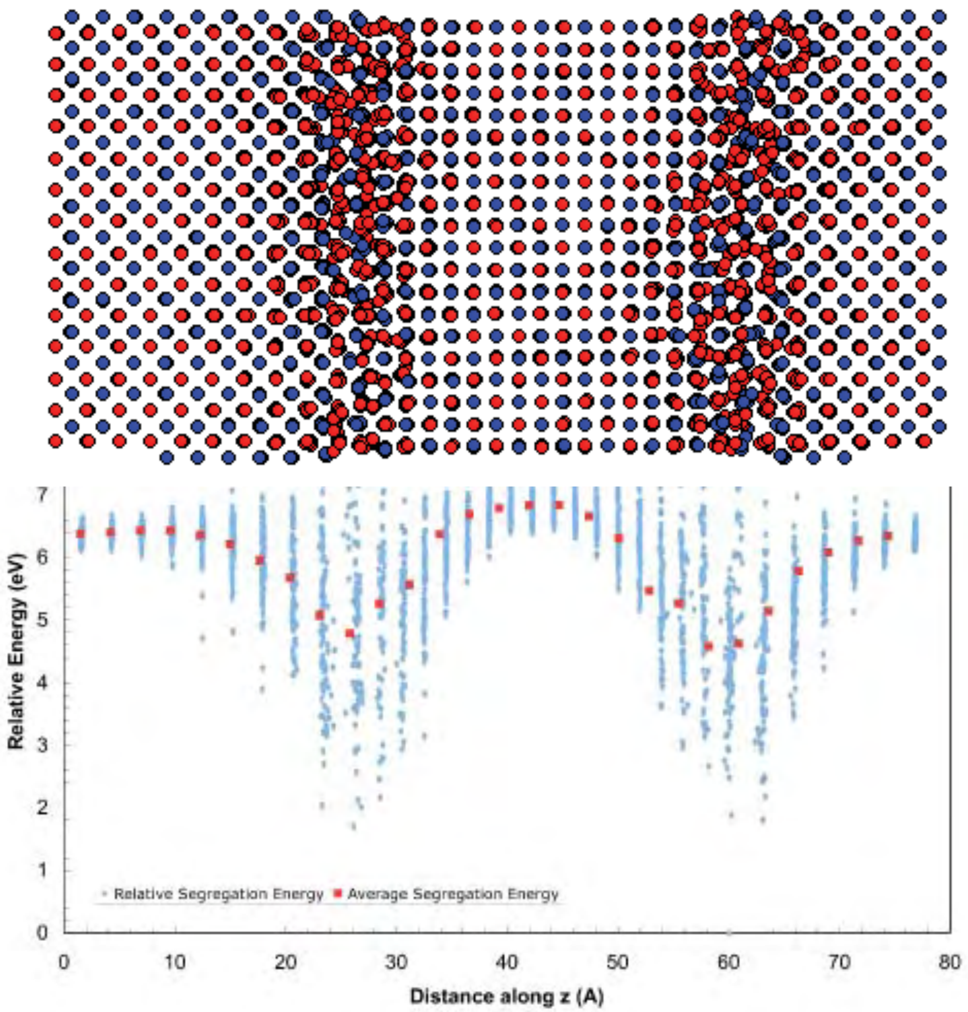


FIG. 1. (Color online) The migration events of the interstitialcy mechanism involving the Xe interstitial atom and two oxygen atoms (labeled O1 and O2). (a) The initial state; (b) the saddle point or transition state; and (c) the final state. The large dark (blue) ball is Xe atom. The dark (red) balls are oxygen atoms. The light (gray) balls are uranium atoms. Periodic boundaries were used in simulations, as indicated by the three-dimensional box around the supercell.

X.-Y. Liu,B.P. Uberuaga, D. A. Andersson, C.R. Stanek, and K.E. Sickafus, "Mechanism for transient migration of xenon in UO_2," *Applied Physics Letters*, **98** (2011) 151902.

Results Part 2: Radiation Produced Defects

Using Pair Potentials for Xe Grain Boundary Segregation



Modern simulation capability allows for consideration of **relevant** microstructural features – e.g. grain boundaries.

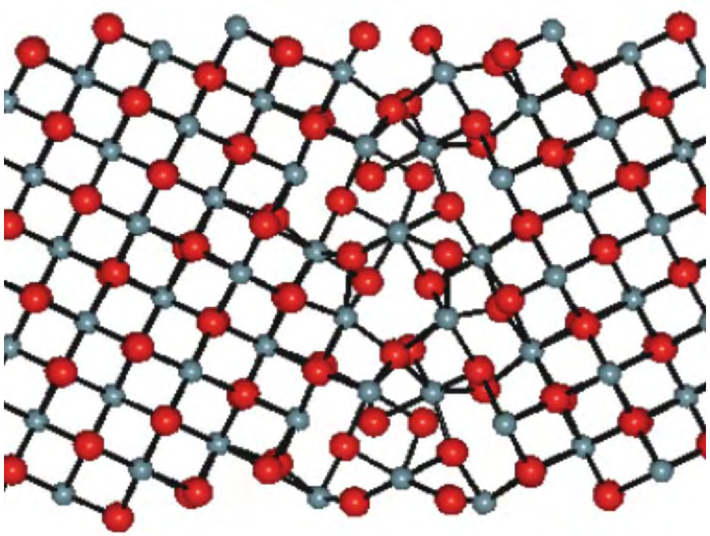
Recent results demonstrate that the atomistic structure of grain boundaries impacts fission gas segregation.

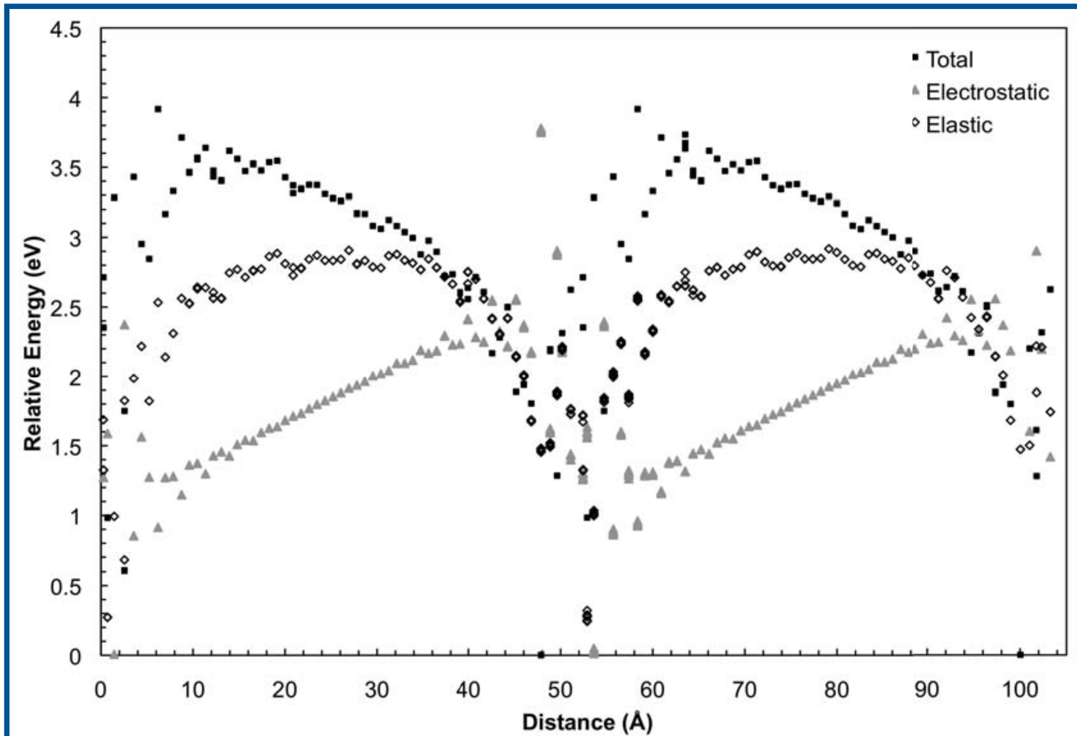
Results at left are for an amorphous boundary – also results for Σ3, Σ5 tilt and Σ5 twist.

"Xenon Segregation to Dislocations and Grain Boundaries," P.V. Nerikar, D. Parftt, D.A. Andersson, S.B. Sinnott, R.W. Grimes, B.P. Uberuaga and C.R. Stanek, submitted to *Physical Review B.*

Results Part 2: Radiation Produced Defects
Grain Boundary Space Charge?

For example, the relaxed structure of Σ5 tilt boundary leads to electrostatic effects that extend much further in to the bulk than other boundary types.



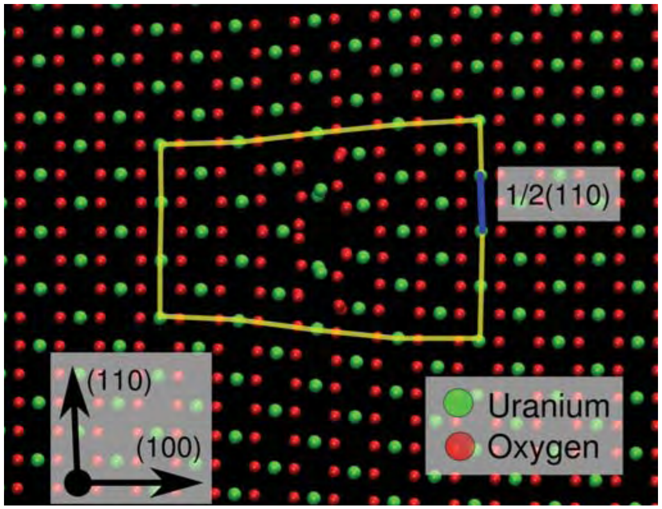


P. Nerikar, C. R. Stanek, S. R. Phillpot, S. B. Sinnott, and B. P. Uberuaga, “Intrinsic electrostatic effects in nanostructured ceramics,” *Phys. Rev. B* **81** (2010) 064111.

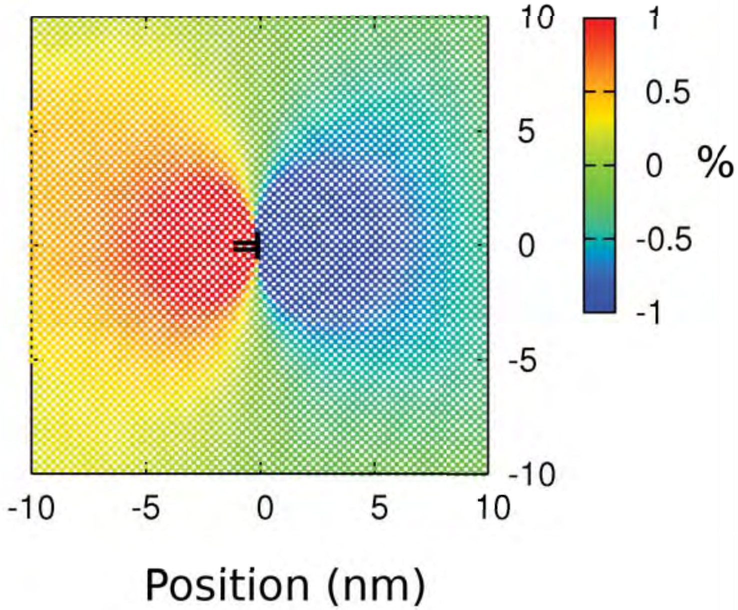
Example of interaction with BES

Results Part 2: Radiation Produced Defects
Collaboration w/ Imperial College: Dislocations

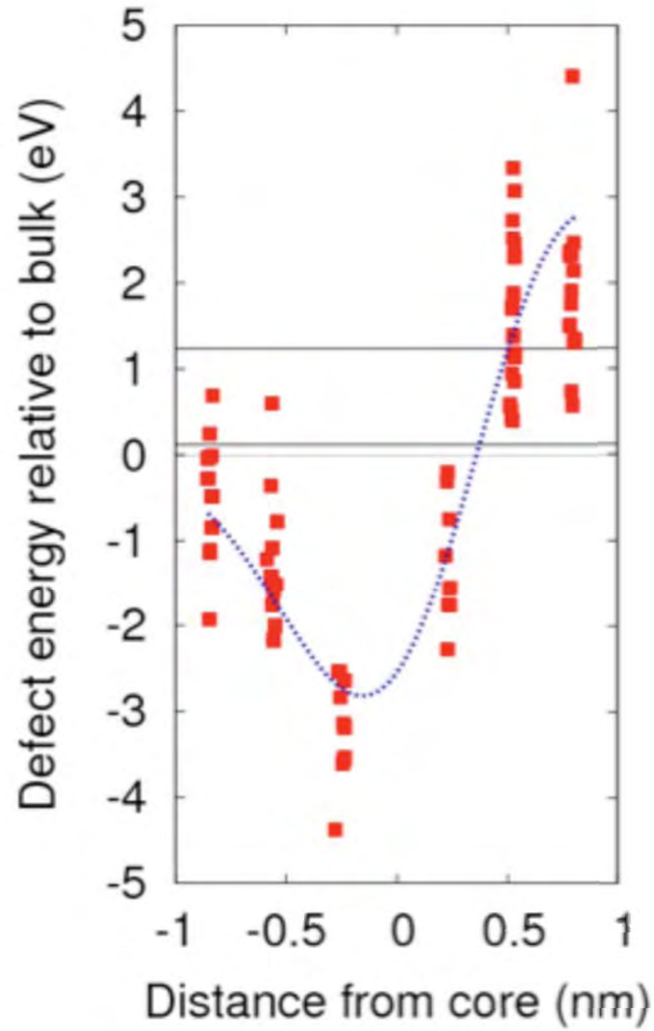
MD simulations of dislocation structure



Corresponding strain field (potentially important for mesoscale simulations)



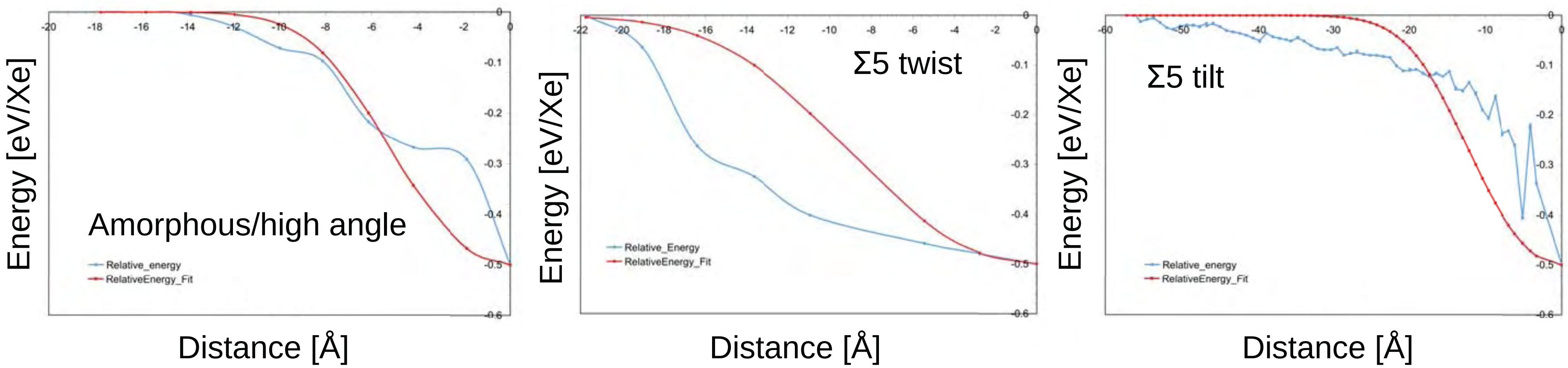
Segregation of Xe near dislocation



Results Part 2: Radiation Produced Defects

Coarse Graining Atomistics for Continuum Calculations

Normalized Xe segregation energies from pair potentials to generate fission gas – GB interaction terms as a function of GB structure



$$f(r) = 1\Big/\left(1+\exp\left(-(r-r_0)^2/k^2\right)\right)-1$$

$$E_m(r) = 2C_1 f(r)$$

$$\boxed{E_m^{gb}(x,r) = xE_m(r)}$$

GB type	k (nm)	C_1 (eV)
∑5 Tilt	1.225	4.09
∑5 Twist	0.922	0.97
Amorphous	0.469	6.42

- "Simplistic" approach designed to capture sink strengths and interaction range.

Results Part 2: Radiation Produced Defects

Coarse Graining Atomistics for Continuum Calculations

- Thermodynamics:

$$G_m = y_{Xe}\,{}^{o}G_{Xe} + y_U\,{}^{o}G_U + y_{Va}\,{}^{o}G_{Va} + RT\left(y_{Xe}\ln\left(y_{Xe}\right) + y_U\ln\left(y_U\right) + y_{Va}\ln\left(y_{Va}\right)\right) +$$

$$+E^{excess} + \sum_i 2y_{Xe}\left(C_i - g_i\left(y_{Xe}\right)\right)\sum_j f_{ij}\left(r_{ij}\right)$$

- Kinetics:

$$M_{XeVa}y_{Va} = \frac{D_0}{k_B T}\exp\left(-\frac{\Delta Q}{k_B T}\right) \quad \Delta Q = \Delta Q_0 - \sum_i\sum_j q_i f_{ij}\left(r_{ij}\right) \quad \Delta Q_0 = E_a^{Xe} = E_{VU}^{F} - E_B + E_m^{VU,C}\left(T\right)$$

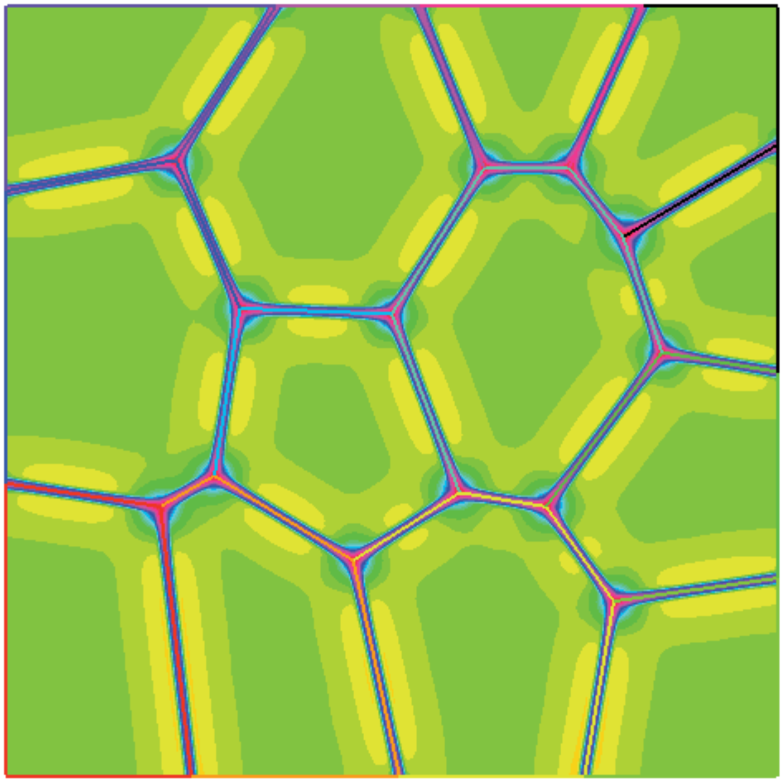
- Thermodynamics + kinetics = Transport equations (μ_{Va}=0)

$$\frac{1}{V_m}\frac{\partial y_{Xe}}{\partial t} = \nabla\cdot\left(\frac{D_0}{V_m}\exp\left(-\frac{\Delta Q}{k_B T}\right)\nabla y_{Xe}\right) +$$

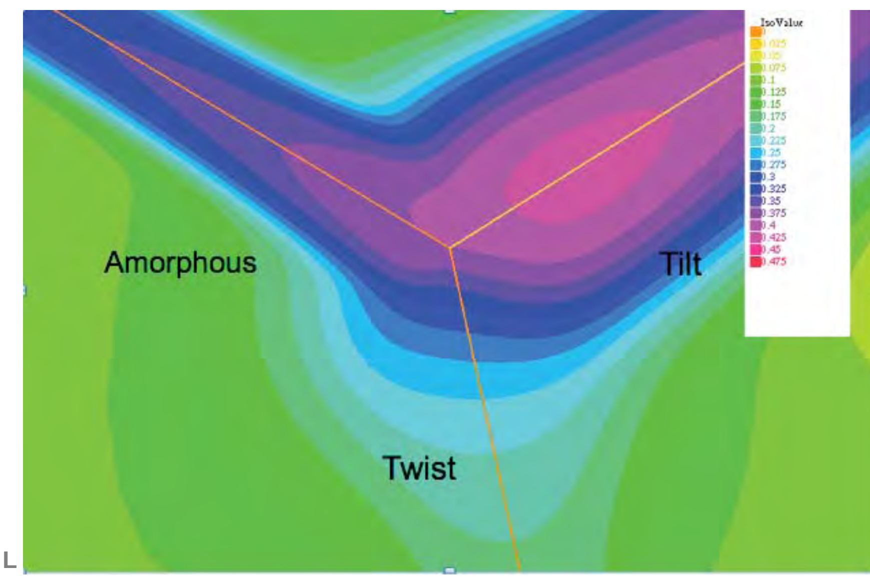
$$+\nabla\cdot\left(\frac{1}{k_B T}\frac{D_0}{V_m}y_{Xe}\exp\left(-\frac{\Delta Q}{k_B T}\right)\nabla\left(\sum_i 2\left(C_i - g_i\left(y_{Xe}\right)\right)\sum_j f_{ij}\left(r_{ij}\right) + \sum_i 2y_{Xe}\left(-\frac{\partial g_i\left(y_{Xe}\right)}{\partial y_{Xe}}\right)\sum_j f_{ij}\left(r_{ij}\right)\right)\right) +$$

$$+\eta\left(t\right)$$

Results Part 2: Radiation Produced Defects
Coarse Graining Atomistics for Continuum Calculations



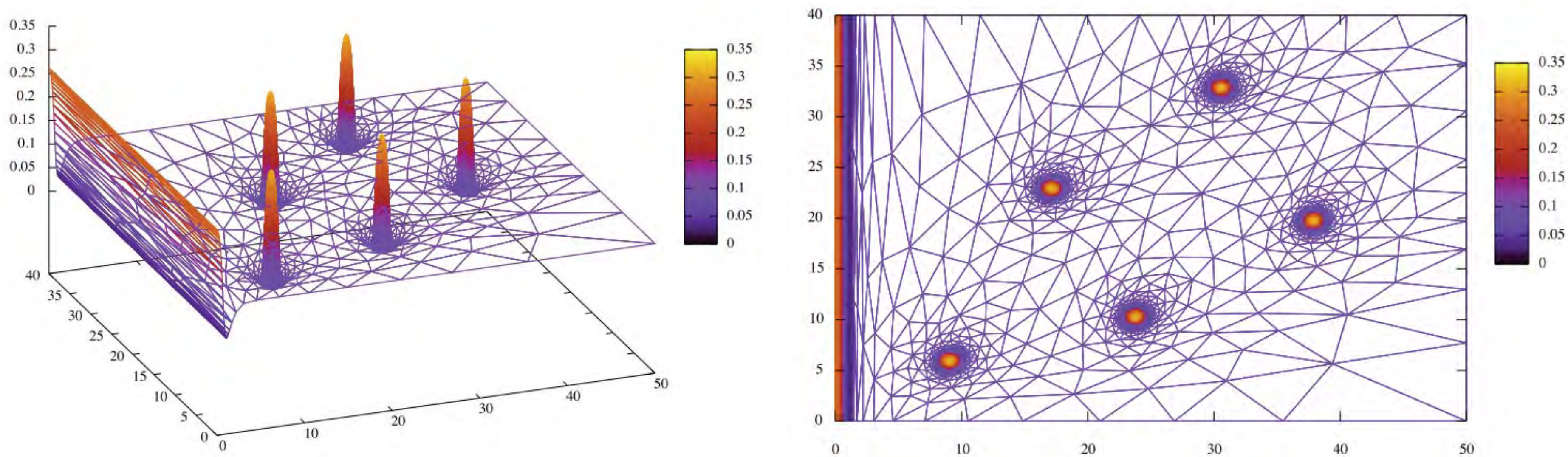
- The larger interaction range of Σ5 tilt boundaries is clearly visible.
- Also the difference in Xe concentration at different boundaries is evident.
- May induce preferential gas bubble nucleation.



Results Part 2: Radiation Produced Defects

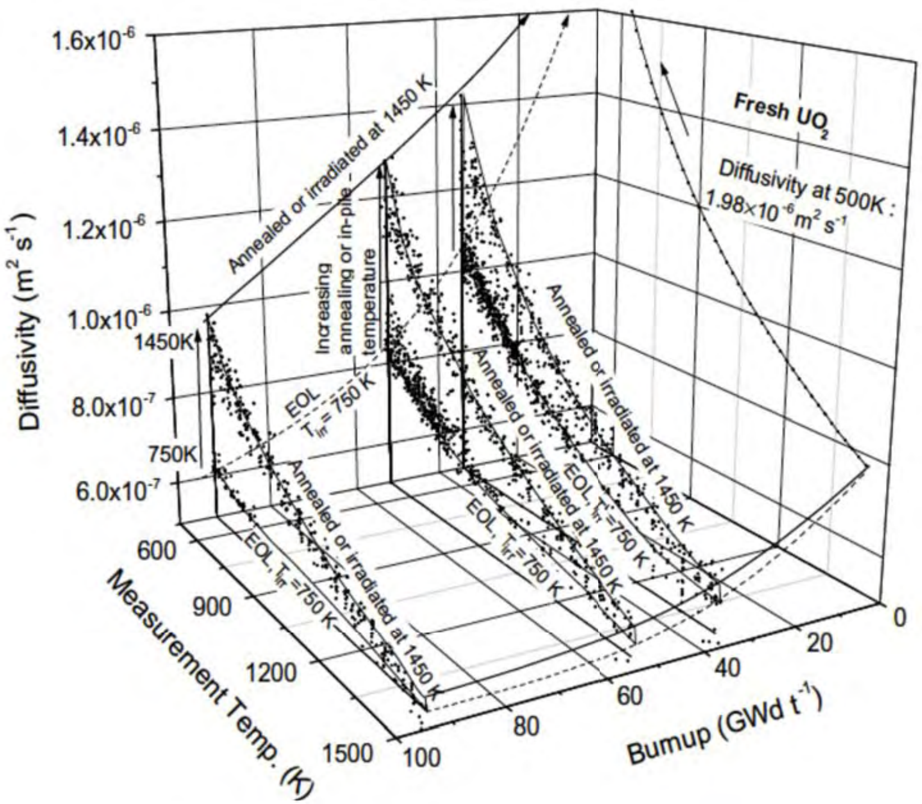
Coarse Graining Atomistics for Continuum Calculations

- Continuum formulation also allows for inclusion of other microstructural features.
- Representative example below illustrates the competition between grain boundaries and dislocations for single fission gas atoms.



Results Part 2: Radiation Produced Defects
A Pragmatic Approach to Thermal Diffusivity

C. Ronchi, *et al*., *J. Nucl. Mater.* **58** (2004) 327



P.G. Lucuta *et al*., *J. Nucl. Mater.* **232** (1996) 166

$$k = k_0\ \kappa_1(\beta)\ \kappa_2(p)\ \kappa_3(x)\ \kappa_4(\tau)$$

where:

k_0 is the unirradiated thermal conductivity

$\kappa_1(\beta)$ is the burnup (fission product) factor

$\kappa_2(p)$ is the porosity/bubble contribution

$\kappa_3(x)$ accounts for the O/M composition

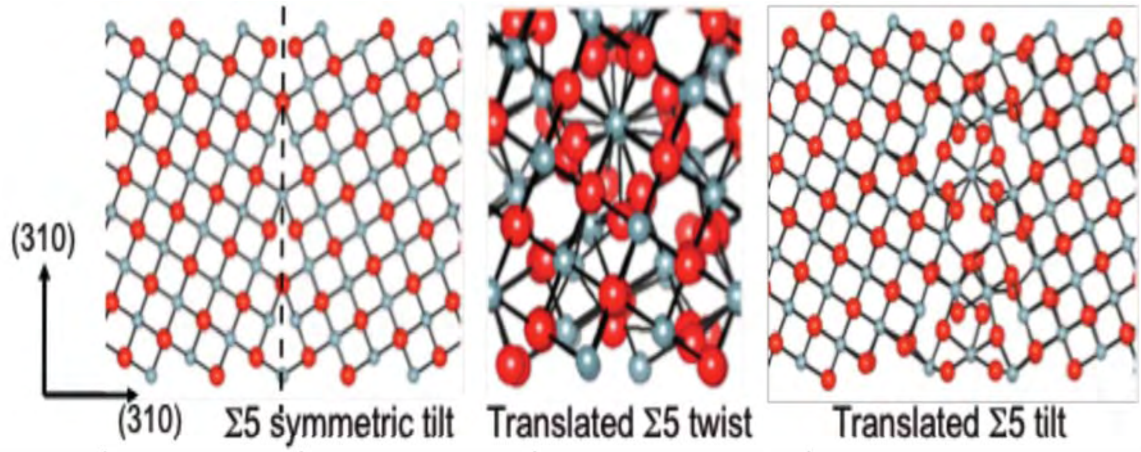
$\kappa_4(\tau)$ refers to radiation damage from neutrons, α-decay and fission

Results Part 2: Radiation Produced Defects

Atomistic Results of UO_2 Kapitza Resistance

Preliminary results of the role of boundary on thermal diffusivity

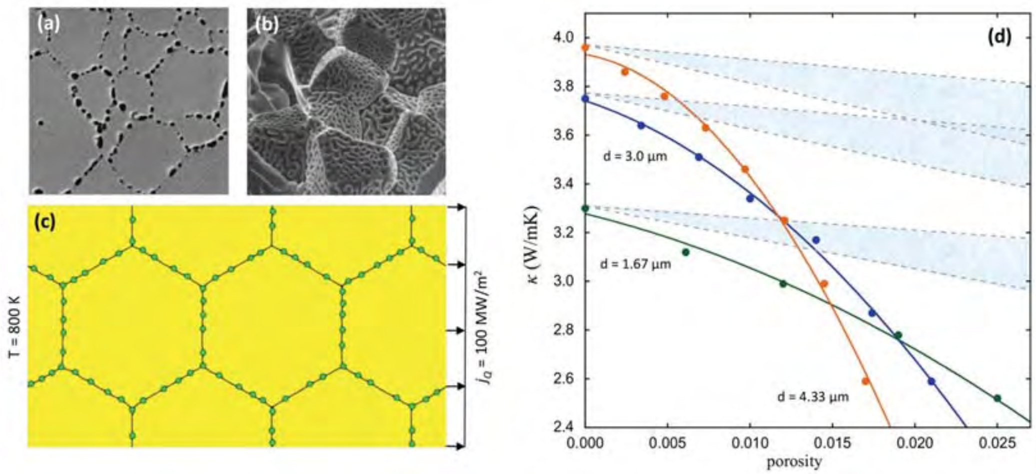
(Kapitza Resistance = measure of an interface's resistance to thermal flow)



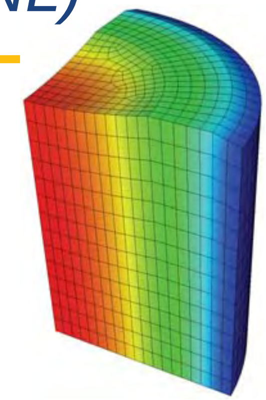
Boundary type	Grain size (nm)	K (W/mK)	Kapitza resistance (m^2K/W)	Kapitza length (nm)
Σ5 (310) symmetric tilt	21.6	7.62	2.05×10^{-9}	56
Σ5 (310) translated tilt	21.6	14.22	0.73×10^{-9}	20.13
Σ5 36.7° translated twist	20.24	9.38	1.00×10^{-9}	39

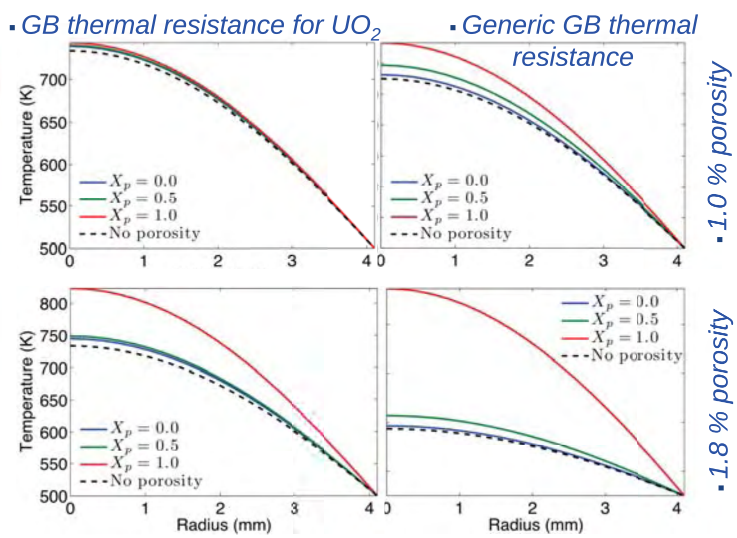
Results Part 2: Radiation Produced Defects

Extension of Atomistics to Mesoscale (INL)



Mesoscale model of thermal conductivity in UO_2 with intergranular bubbles as a function of grain size d and porosity using the GB thermal resistance from MD





Lower length scale information can have a significant impact on macroscale fuel performance

Radial temperature profiles in a UO_2 fuel pellet, where the fraction of intergranular bubbles X_p is varied from 0 to 1

Summary – Future Work

- Atomistic simulation results shown for natural defects, mass and thermal transport in UO_2, and initial upscaling steps have been taken.

For future experiments, focus on the role of:

* composition/nonstoichiometry - thermodynamics (calorimetry, gravimetry, etc), structure (XRD, neutrons for oxides, light sources, etc.

* microstructure (SEM/EBSD, HR-TEM, etc.),

*and rad effects (ion beams, reactors) – keeping in mind that this affects the first 2, and also limits what we can measure.

On:

* Mass (FG) transport

* Thermal transport (laser flash, local measurements, etc.)

Development of experimental techniques to augment theory and simulation of thermal transport in oxide nuclear fuel

A.T. Nelson (MST-7)

Recent years have seen considerable advances in the capabilities of computational techniques to address many of the challenges inherent to operation of nuclear fuels. However, the new challenge for experimentalists is the development of novel techniques and representative material systems able to provide both requisite fundamental data sets as well as benchmark predicted results. The greatest immediate challenge is the need to differentiate and produce microstructures as a function of their most basic components in order to understand the role of specific grain boundary types, porosity morphology, and defect chemistry on one of the most critical aspects of oxide fuel operation, thermal transport.

A novel experimental approach has been undertaken to address these most basic components that govern heat transport in nuclear fuels both prior to and during irradiation. The first challenge is development of a thermophysical property measurement infrastructure with the precise controls necessary to execute the necessary measurements across the wide temperature range of importance to reactor operation. Second, material samples must be prepared in order to isolate, maintain, and address the effects of individual microstructural aspects on the thermophysical properties of interest. Examples of methods currently employed to characterize the impact of defect chemistry and grain boundary character on the thermal conductivity of uranium dioxide will be discussed. Finally, extension of this approach to incorporate effects such as radiation damage and fission products encountered during burnup is planned through both rabbit irradiations as well as other novel sample preparation techniques.

Development of Experimental Techniques to Augment Theory and Simulation of Thermal Transport in Oxide Nuclear Fuels

Andy Nelson, Darrin Byler, John Dunwoody, Erik Luther, Jack Henderson, Pedro Peralta, and Ken McClellan

Materials Science and Technology Division

UNCLASSIFIED

Presentation Outline

- **Orientation to ceramic fuel systems of study**
- **Addressing the experimental challenge provided by modeling and simulation through Separate Effects**
- **Thermophysical property characterization**
- **Separate Effects approach to thermal transport in ceramic fuels**
  - Single crystal growth
  - O/M control
  - Path for study of microstructure effects
- **Incorporation of irradiation campaigns into a Separate Effects approach**

Ceramic Nuclear Fuels at LANL: Systems of Experimental Emphasis

APPLICATION: *Existing Fleet* | *New Builds* | *Advanced Fuel Cycle*

CERAMIC FUEL SYSTEM

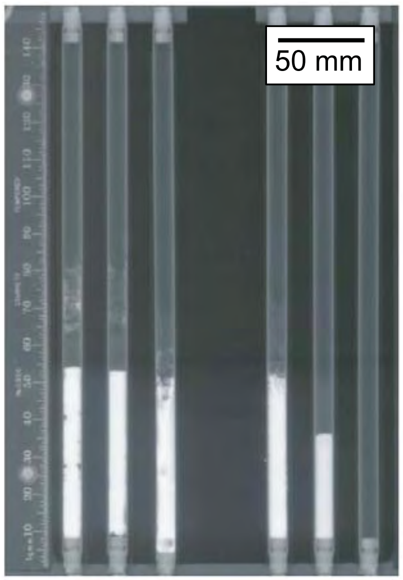
- UO_2
- *CONVENTIONAL MOX $(U,Pu)O_2$*
- ThO_2 $(Th,U)O_2$ $(Th,Pu)O_2$
- *MOX + MINOR ACTINIDES (Np,Am)*
- UN PuN

PLUTONIUM SURROGATES

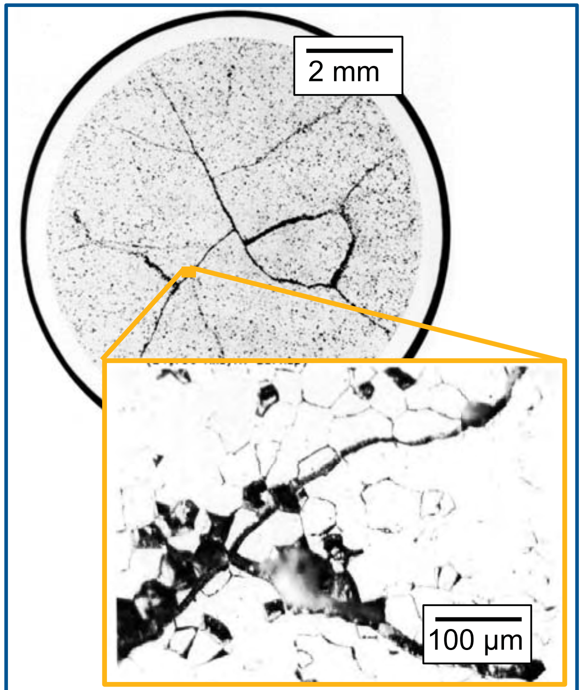
- $(U,Ce)O_2$ $(U,{}^{242}Pu)O_2$
- ZrN

MODELING/THEORY-DRIVEN COMPOSITIONS

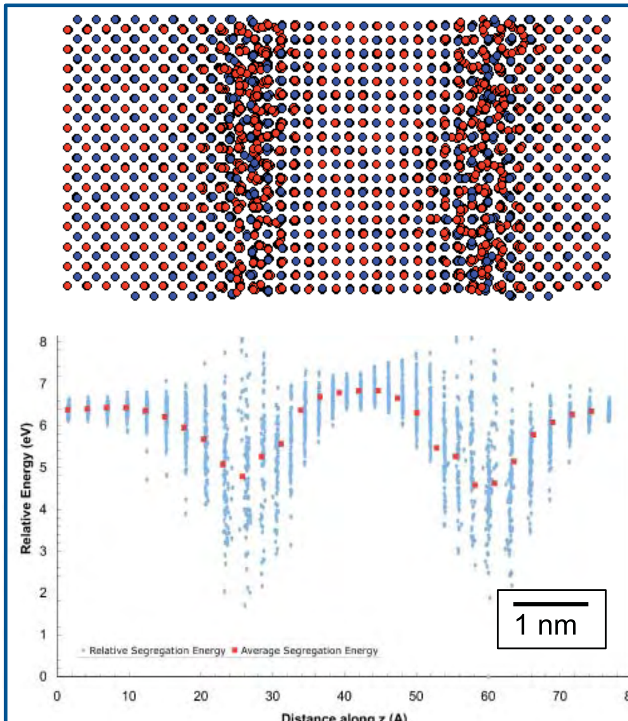
Challenge of Linking Traditional Post Irradiation Examination to Modeling Efforts



Traditional neutron radiography of metal fuels following irradiation



~2% FIMA 95/5 (Th,^{233}U)O_2 (Campbell et al. WAPD-TM-1606 1987)



Xe segregation to grain boundaries (Nerikar et al. submitted to J Nuc Mat)

EXPERIMENTAL CHALLENGE: DEVELOPING TECHNIQUES AND APPROACHES TO BRIDGE THIS GAP

Beyond Traditional Post Irradiation Examination: The "Separate Effects" Approach

SEPARATE EFFECTS — **DESIGN EXPERIMENTS TO ISOLATE SPECIFIC PHENOMENA AND CHARACTERIZE THEIR IMPACT ON FUEL PROPERTIES AND BEHAVIOR**

FABRICATION OF "SYNTHETIC" HIGH BURNUP STRUCTURES

- Necessary to isolate numerous variables encountered during burnup
- Avoid temporal & radiological concerns
- Facilitates full suite of modern analysis methods (e.g. TEM)

DEVELOPMENT OF NEW TECHNIQUES

- Complete understanding of all actors and effects in fresh fuel is vital
- Must include unprecedented controls to account for complex behavior of oxides at high temperatures

EXPANSION OF TRADITIONAL PIE

- Hot cell capabilities are requisite for in-pile irradiations
- Expansion and modernization needed to match precision of fresh fuel analyses

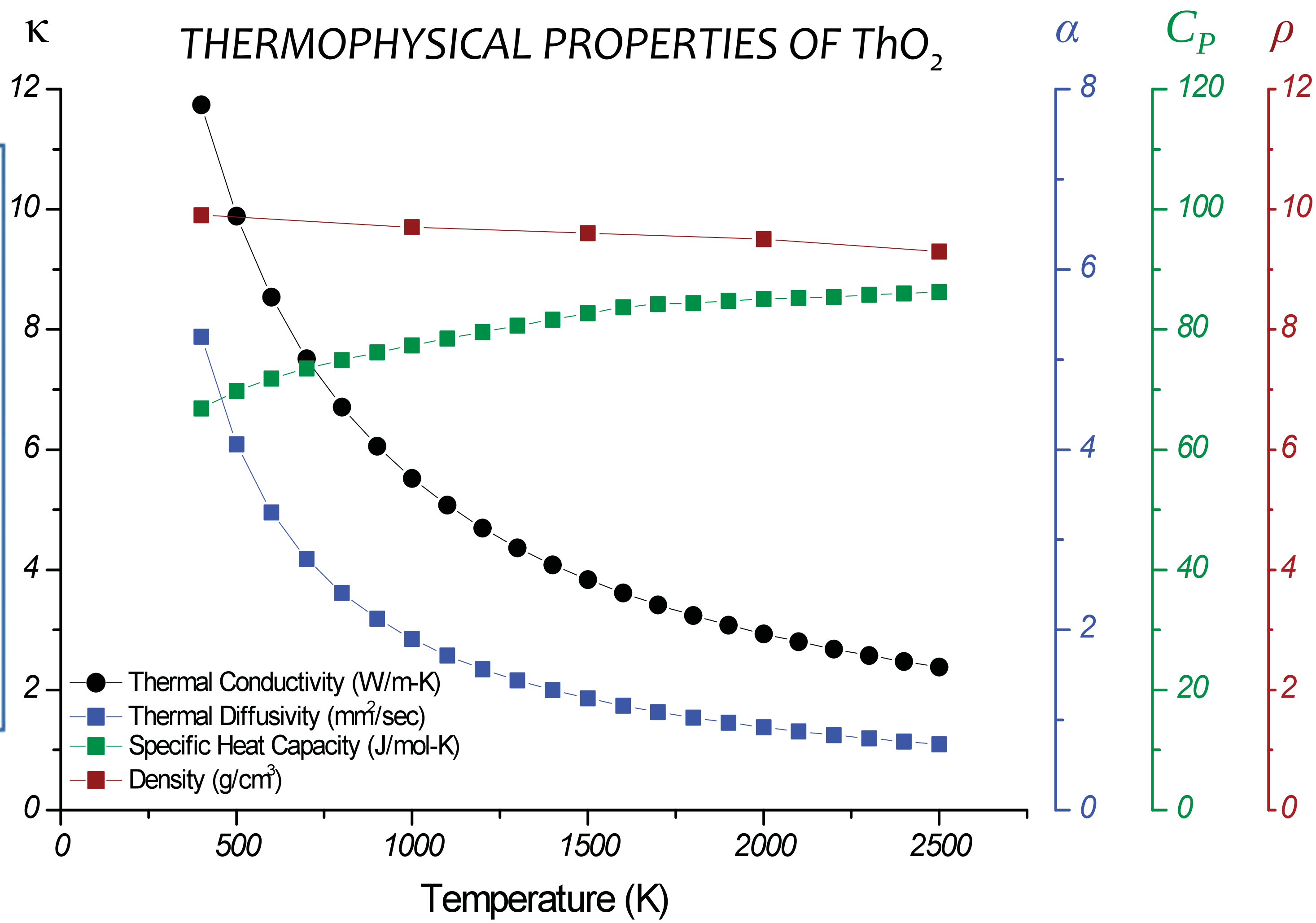
Application of Modeling and Experiment to Study of Thermal Transport in Ceramic Fuels

- **Thermal transport dominates design, operation, and safety analysis in ceramic fuels**
- **Existence of thermal conductivity and melt point correlations as a function of burnup for UO_2 limited to engineering scale applicability**
  - Significant uncertainty exists – even prior to irradiation
  - Translation to other reactor designs, derivative fuel forms e.g. $(Th,U)O_2$ requires return to substantial uncertainties
- **Motivation exists to develop predictive physics-based computational tools and codes describing thermal transport in UO_2**
- **Models are progressing… but an integrated experimental approach has lagged, motivating a Separate Effects approach**

Determination of Thermal Conductivity using Modern Thermophysical Property Techniques

$$\kappa = \alpha \cdot C_P \cdot \rho$$

κ	Thermal Conductivity
α	Thermal Diffusivity
C_P	Specific Heat Capacity
ρ	Density



Determination of Thermal Conductivity using Modern Thermophysical Property Techniques

α Thermal Diffusivity

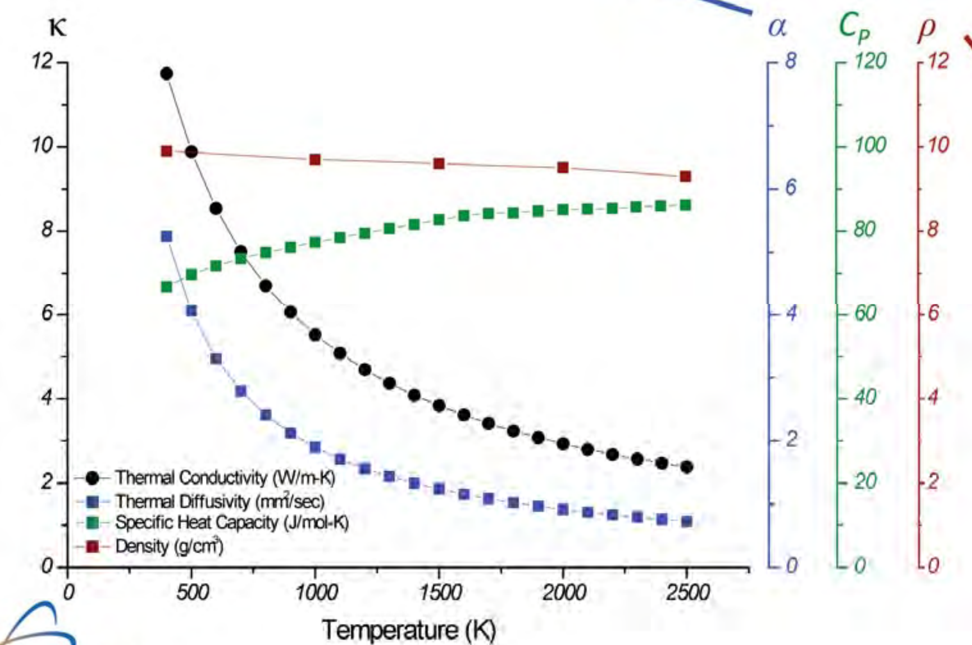
Laser Flash Analysis (LFA)

C_P Specific Heat Capacity

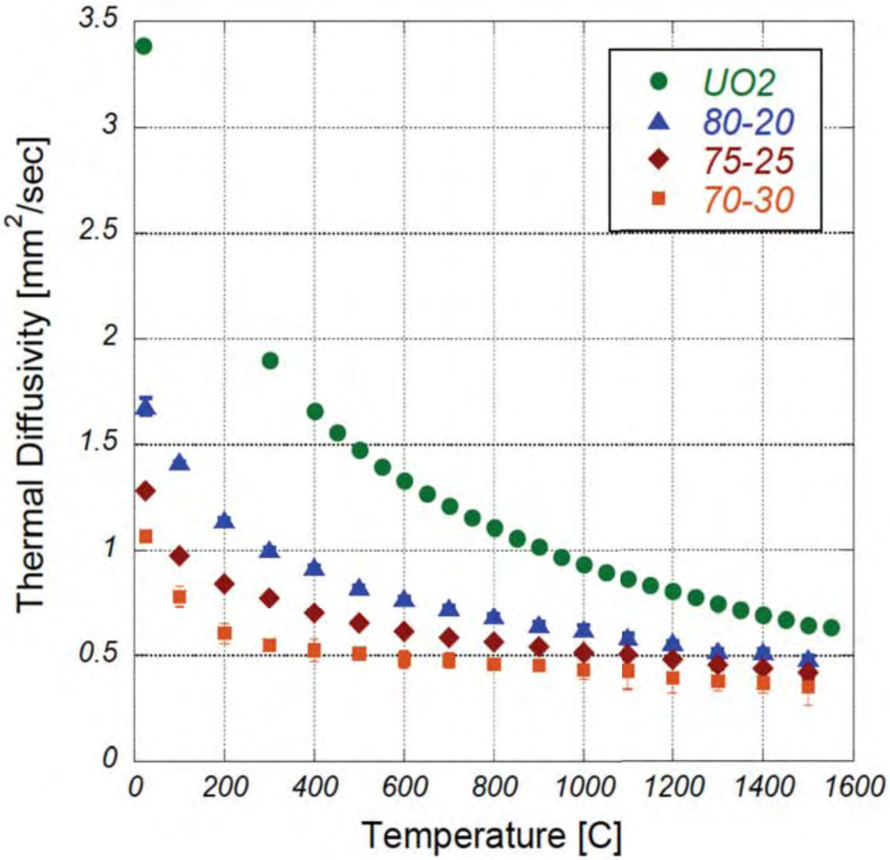
Differential Scanning Calorimetry (DSC)

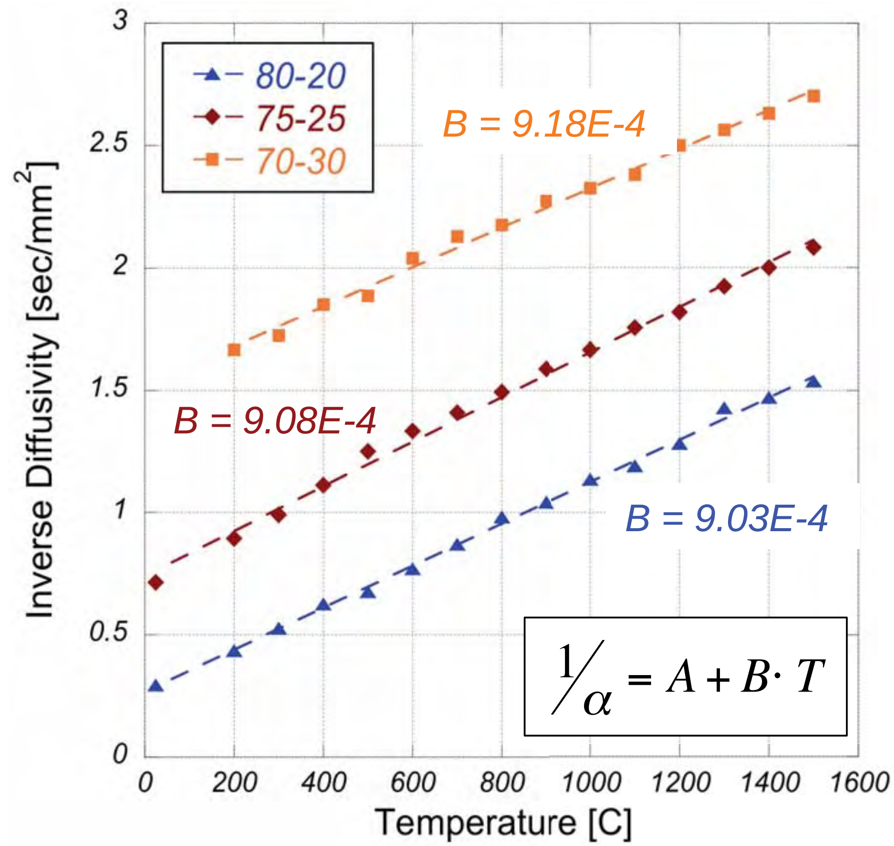
ρ Density

RT density measurement with dilatometry data



Effect of Cerium Content on Thermal Conductivity of $(U,Ce)O_2$

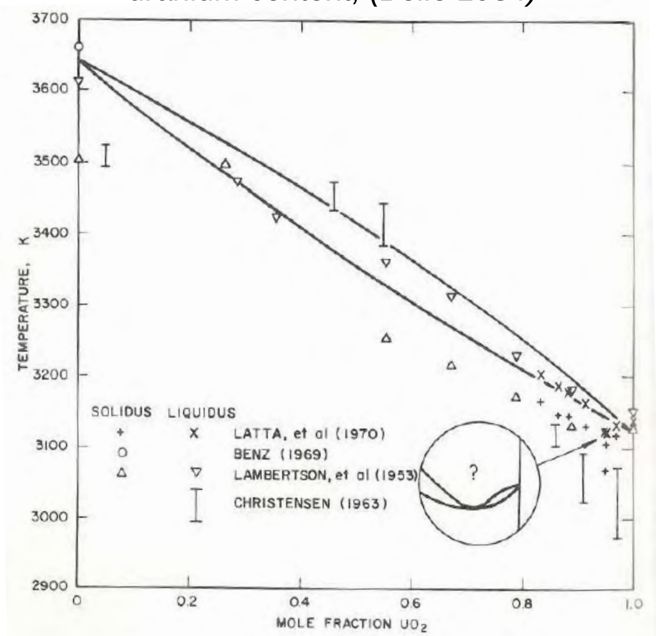




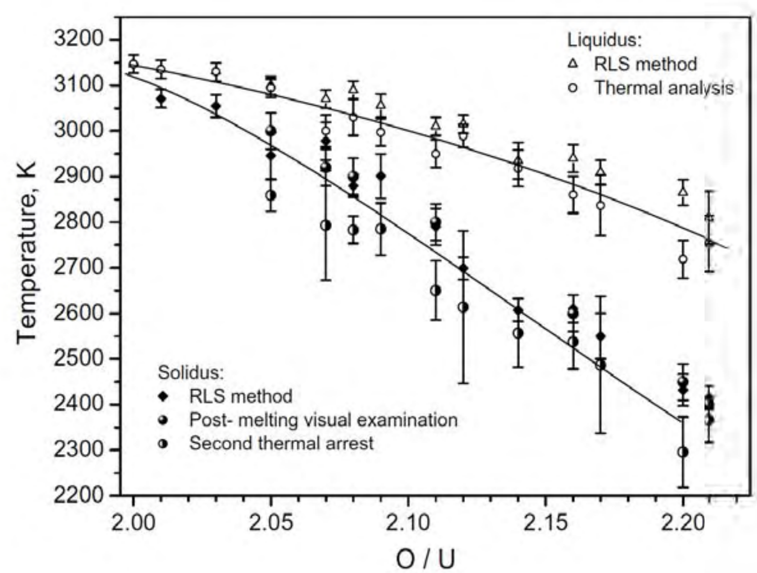
Characterization of $(U,Ce)O_2$ demonstrates classic independence of phonon-phonon scattering (*B*) on composition. Density of lattice cation defects (*A*) is source of degradation.

Uncertainty of Melt Point in Ceramic Fuels

Solidus / Liquidus of (Th,U)O_2 as a function of uranium content; (Belle 1984)



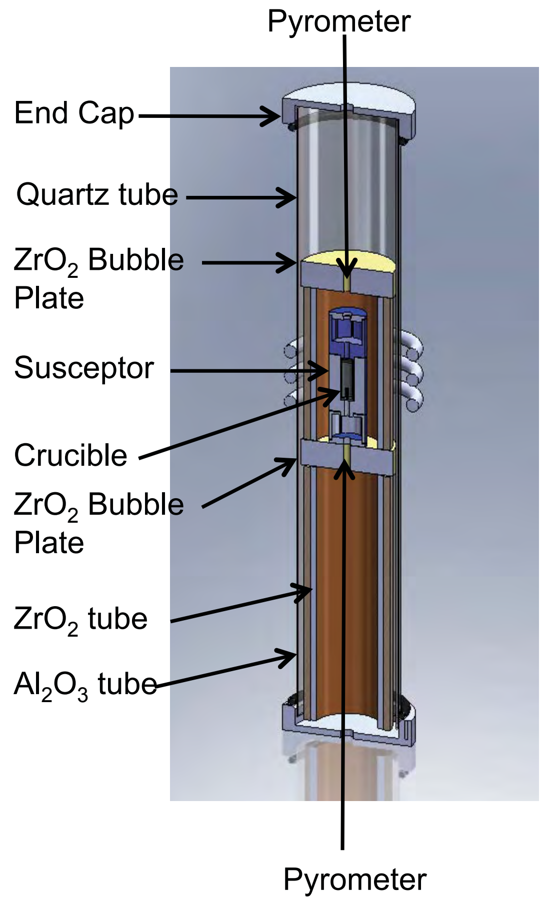
Solidus / Liquidus of hyperstoichiometric UO_{2+X} as a function of X; (Manara J Nuc Mat 2005)

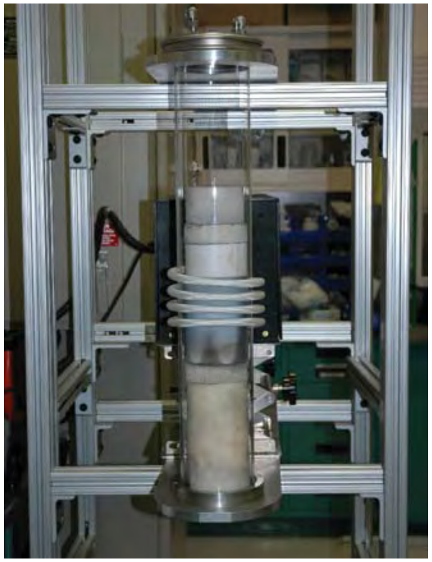


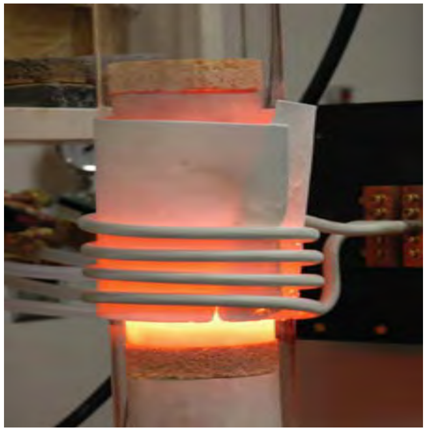
- **Melt point (T_M) drives determination of reactor operational limits**
- **Capability to determine T_M as a function of necessary variables (e.g. composition, burnup) unavailable elsewhere in US**

Development of Melt Point Determination System for Ceramic Fuels

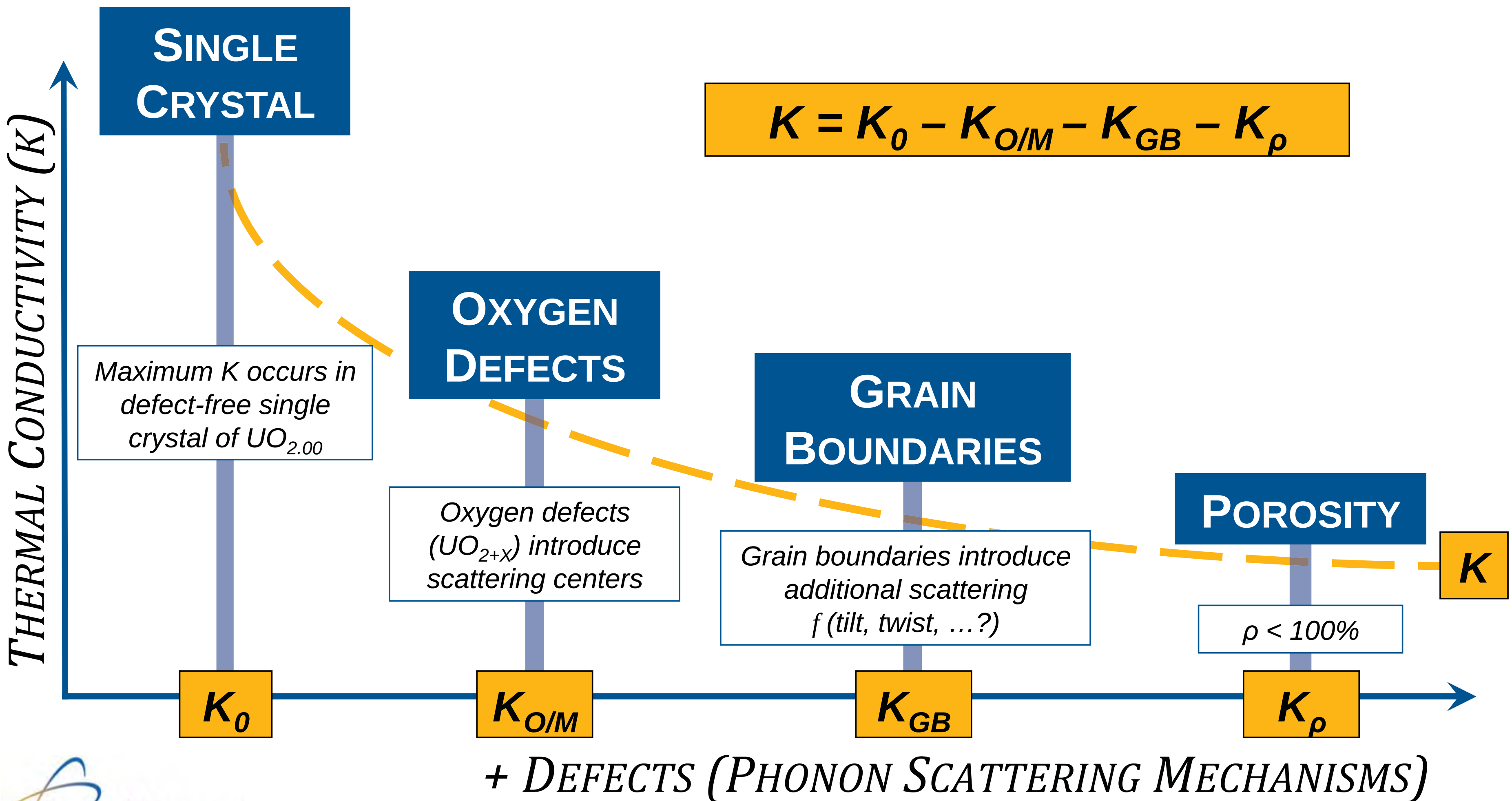
- **System developed based on existing designs but modified to meet safety concerns of fuel analysis**
- **Two color pyrometer monitors tungsten-encased sample heated through RF field direct coupled to graphite susceptor**
- **Calibrated using known ceramic T_M to 2700 ± 20°C (system capable > 3500°C)**
- **Full atmosphere controls allow study of O/M effects on T_M of UO_{2+X}**







Principle Mechanisms Governing Thermal Transport in Single Phase Ceramic Systems



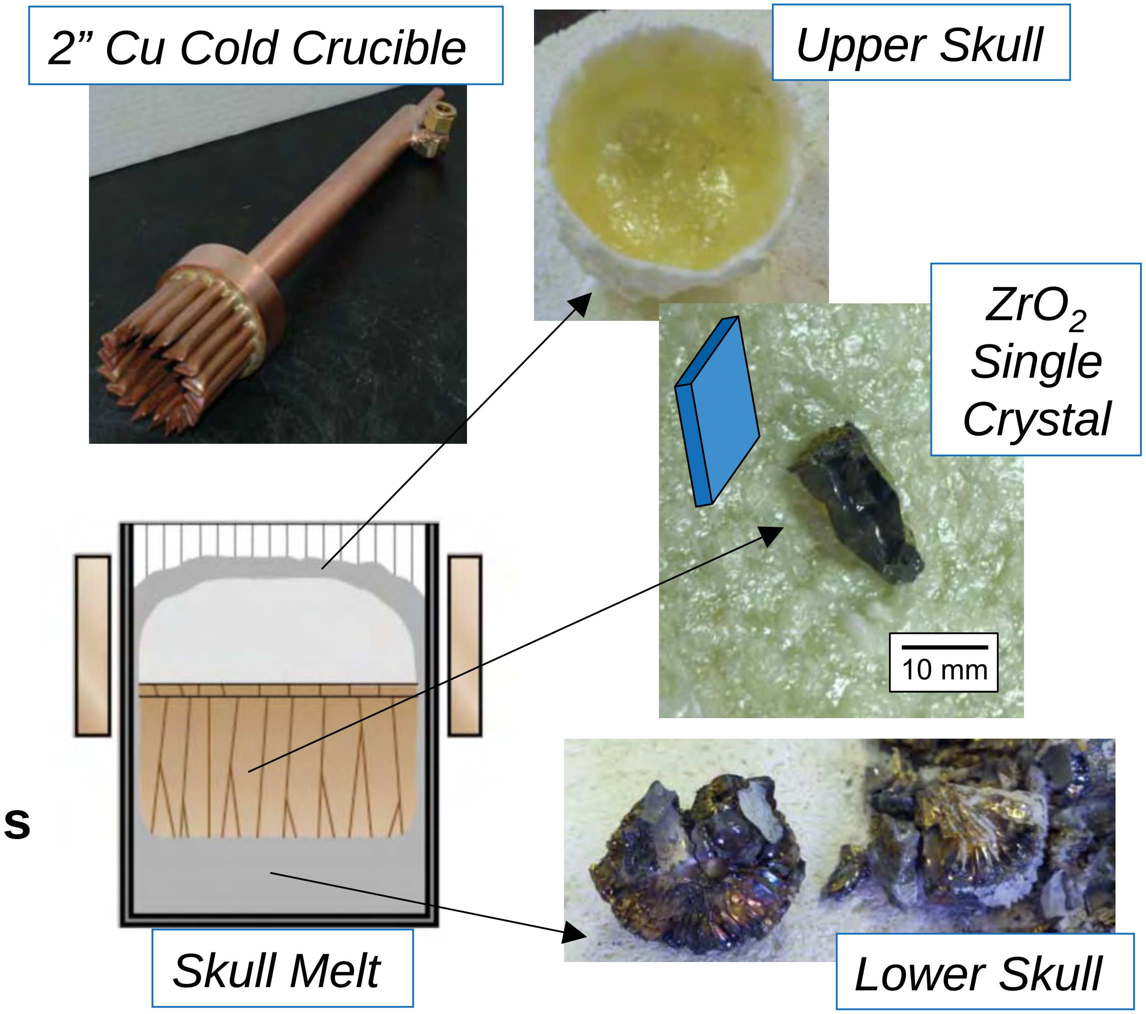
Requirements for Addressing Separate Thermal Transport Effects in UO_2

SEPARATE EFFECT		TECHNIQUE NEED / APPROACH
SINGLE CRYSTAL	K_0	*Capability to fabricate UO_2 single crystals of suitable geometry to measure K_0*
OXYGEN DEFECTS	$K_{O/M}$	*Maintain fixed O/M over wide temperature range within necessary thermophysical property measurement hardware to measure $K_{O/M}$*
GRAIN BOUNDARIES	K_{GB}	*Produce / isolate specific grain boundaries of interest and measure K_{GB} (Kapitza Resistance)*
POROSITY	K_ρ	*Incorporate known porosity morphologies into samples of controlled O/M & GB; validate Synergistic Effects* ***… ONE EFFECT WITH APPRECIABLE EXISTING DATABASE***

Production of Uranium Dioxide Single Crystals

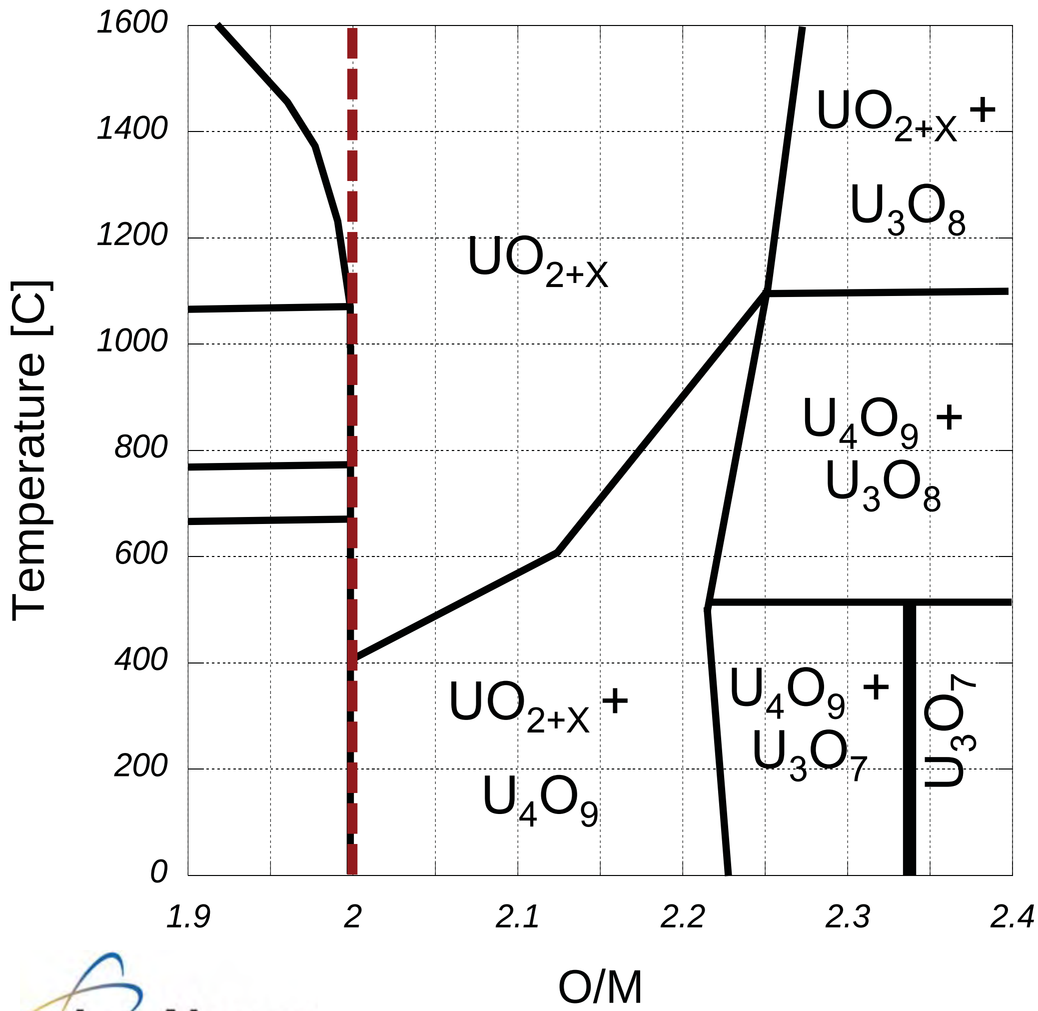
SINGLE CRYSTAL K_0

- **Skull melting chosen due to its ability to incorporate atmosphere control**
- **Previous industrial work has driven design process**
- **Y_2O_3-stabilized ZrO_2 (T_M ~ 2700°C) single crystals grown successfully**
- **UO_2 production begins Summer 2011; $(U,Ce)O_2$ in FY12**



Complexity of Oxidation Behavior in U-O System

OXYGEN DEFECTS $K_{O/M}$

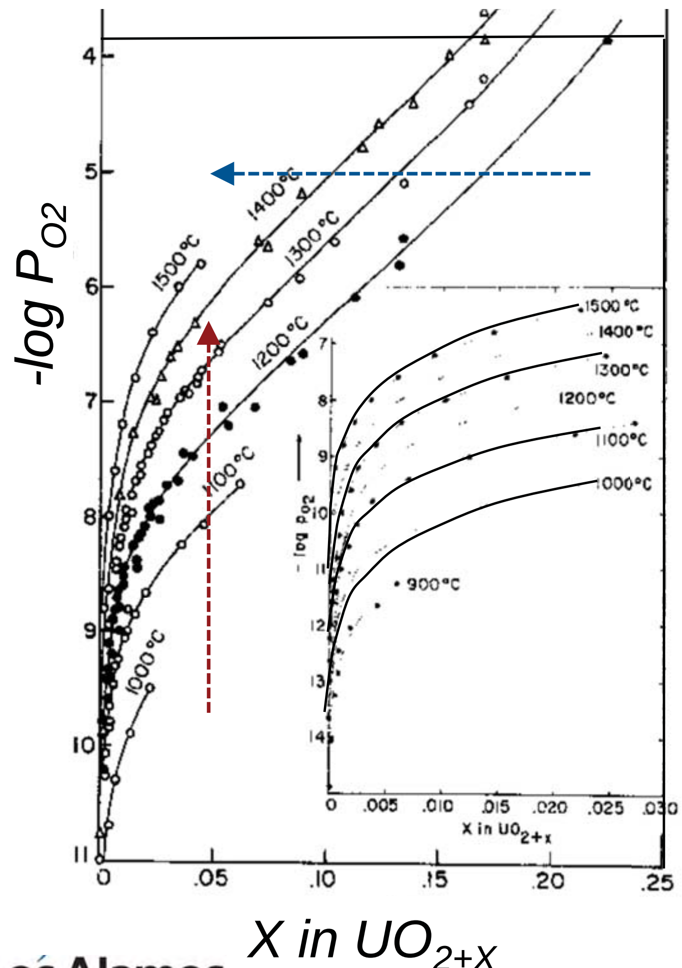


- **Wide fluorite phase field poses significant challenge to characterization of UO_2 properties at temperature**
- **Standard nomenclature is 'O/M' ratio e.g. UO_2 = 2, U_4O_9 = 2.25, U_3O_8 = 2.66**
- **Any experimental studies of UO_2 or derivative systems - e.g. $(Th,U)O_2$ - must be continually aware of this fact… *and its dynamic nature***

Challenge of O/M Control in Experimental Environments

OXYGEN DEFECTS $K_{O/M}$

Hagemar et al., J Inorg. Nucl. Chem ***28*** *(1966)*

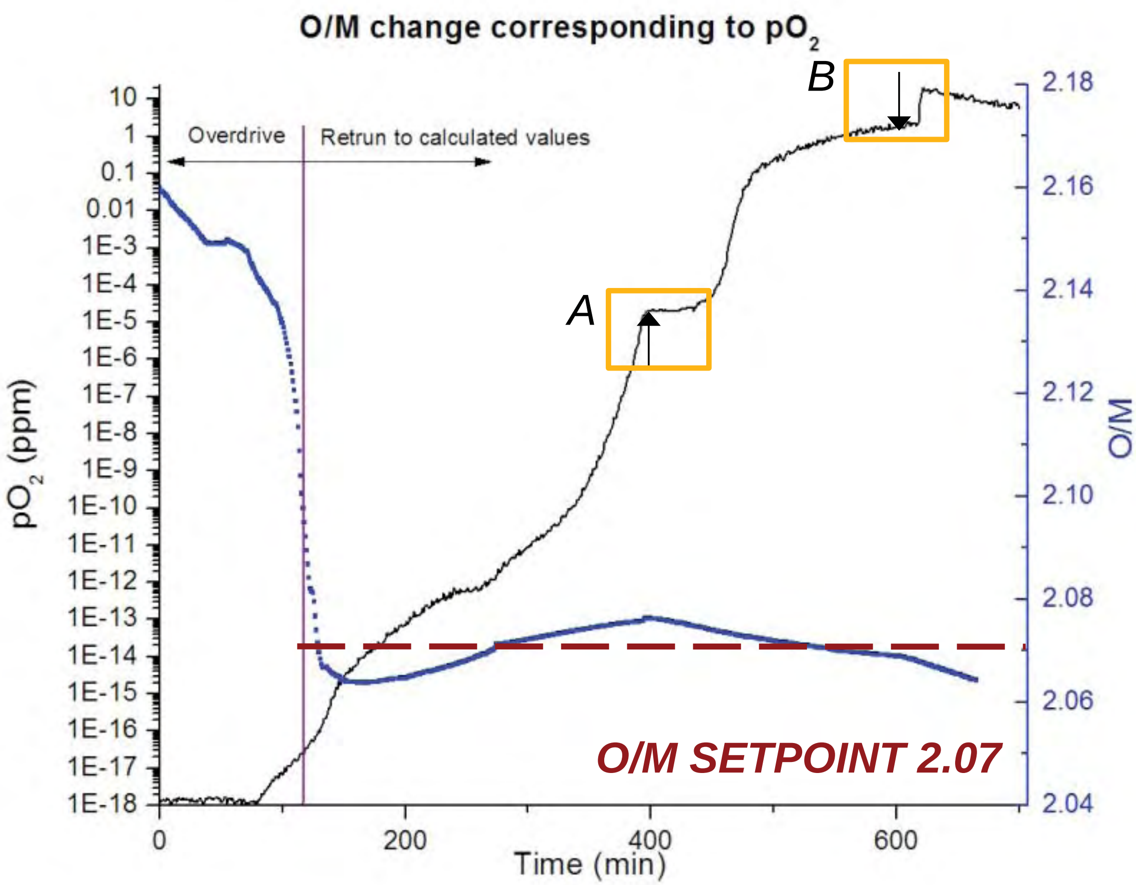


- **Uranium cation charge compensation allows ready accommodation of oxygen defects in fluorite phase field (UO_{2+X})**
- **Fixed gas partial oxygen pressure will force oxygen activity (and therefore O/M) to evolve with temperature with static gas chemistry (blue line)**
- **Solution is use of dynamic gas mixing controls to match input oxygen partial pressure to temperature (red line)**

Implementation of O/M Control in Experimental Environments

OXYGEN DEFECTS $K_{O/M}$

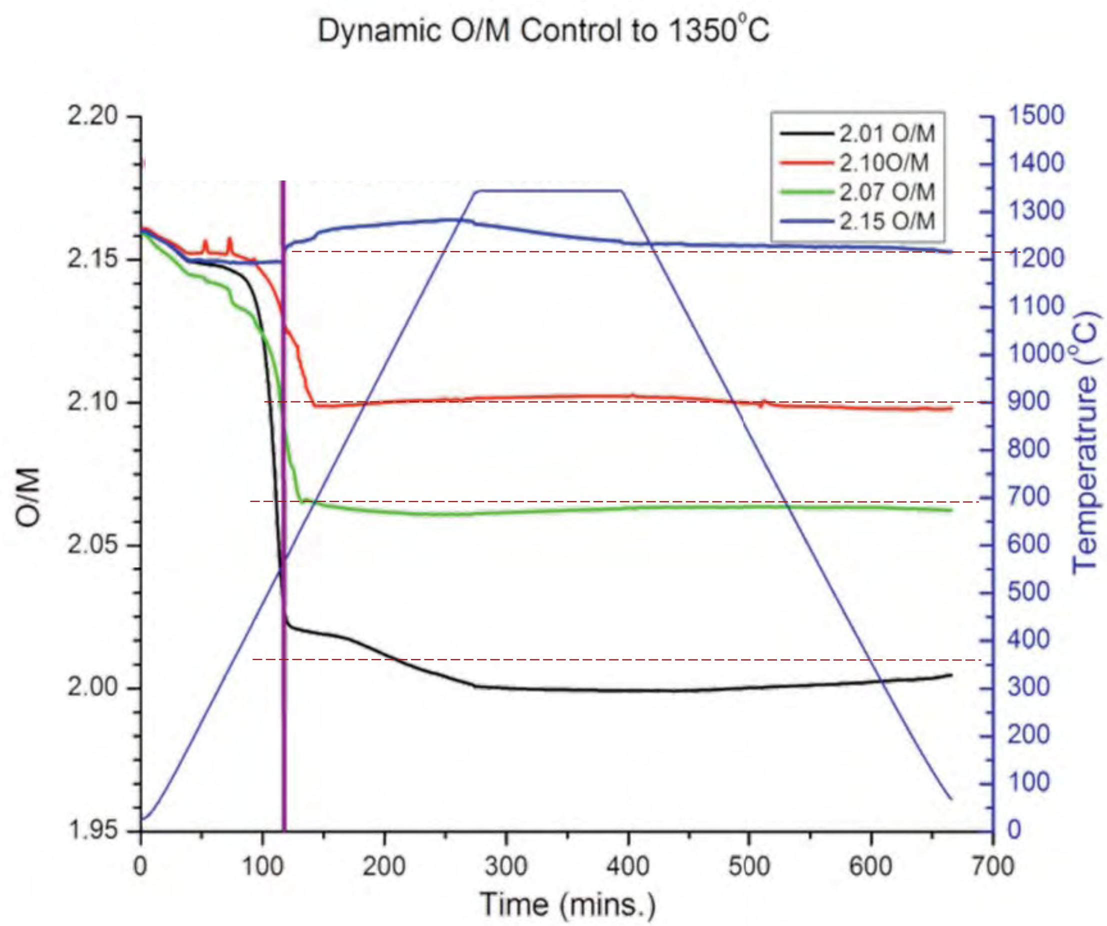
- **Dynamic gas mixing controls allow real time mixing of Ar/Ar-O gas ratios to drive system P_{O2} to desired region to maintain specific O/M**
- **Pellets mass monitored using TGA reveals dynamic O/M during temperature profile (1350°C at right)**



Implementation of O/M Control in Experimental Environments

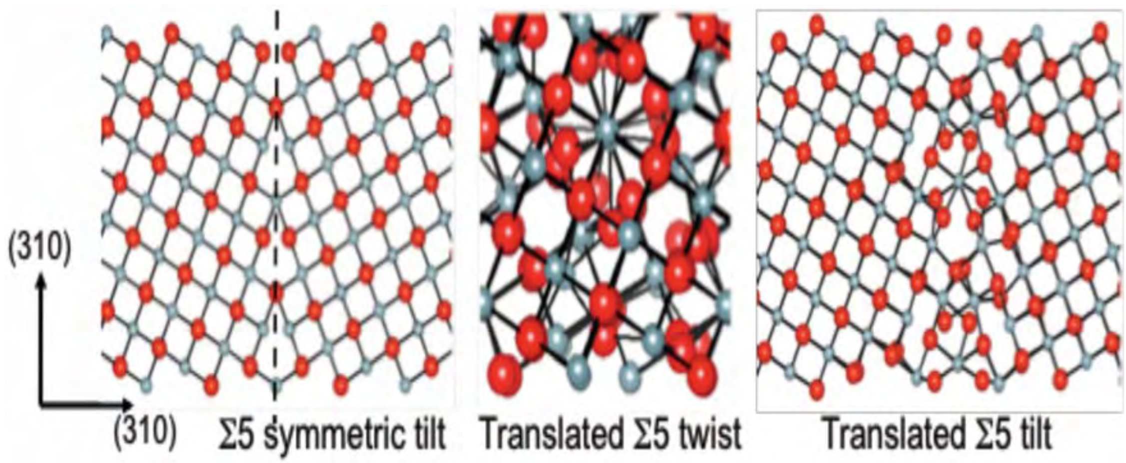
OXYGEN DEFECTS $K_{O/M}$

- **Goal is to maintain fixed O/M ±0.01 throughout entire temperature range of interest (RT – 1600°C)**
- **Implementation of gas mixing / control system in dilatometer, DSC and LFA provides path toward unprecedented UO_2, $(U,Ce)O_2$ and $(Th,U)O_2$ fabrication and property studies (with proper thermochemical models)**

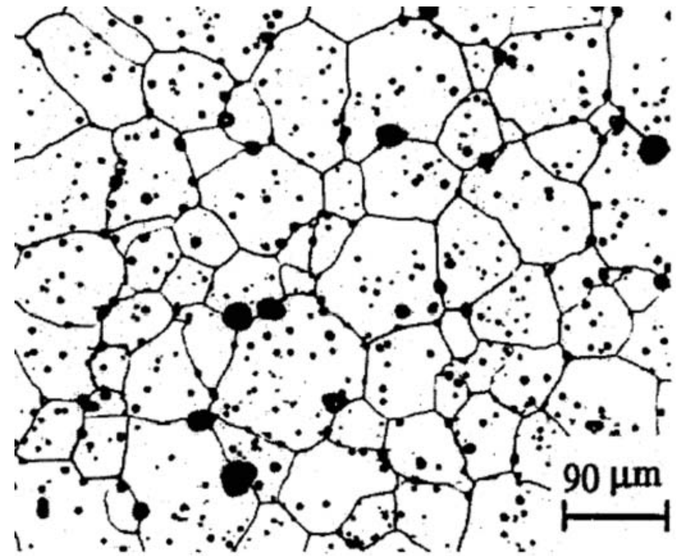


Challenge of Addressing Grain Boundary Effects

GRAIN BOUNDARIES K_{GB}



Grain boundary misorientations constructed for calculation of Kaptiza resistance



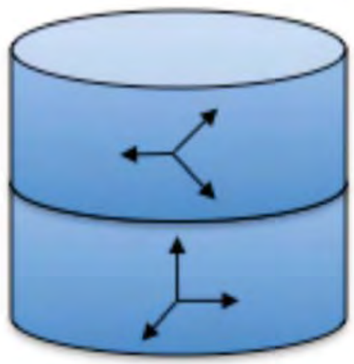
*Conventional UO_2 grain growth study micrograph (Song J Nuc Mat **209** 1994)*

- **Modeling of heat flow across specific grain boundary misorientations yields Kapitza resistance**
- **Challenge is development of techniques that can benchmark these values and incorporate them into real microstructure**

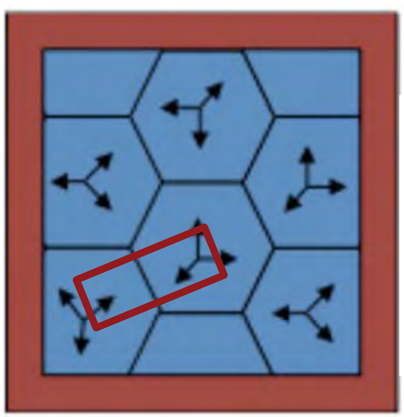
Measurement of Grain Boundary Effects: Path Forward

GRAIN BOUNDARIES K_{GB}

OPTIONS FOR SAMPLE FABRICATION



A. Produce desired boundary through two single crystals



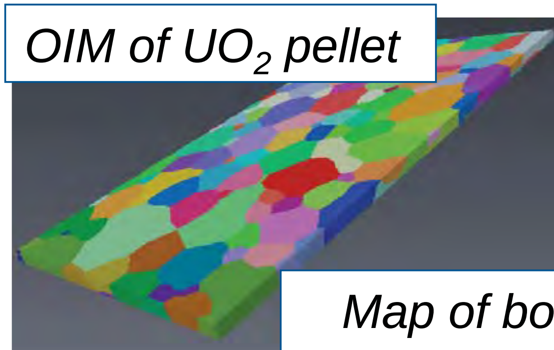
*B. Analyze texture of large grained sample, harvest boundary of interest **(need ~ mm of material)***

- **Most basic & vital parameter is Kapitza resistance determined for a single grain boundary**
- **Sample preparation is greatest challenge, but options exist for conventional measurement (LFA)**
  - Artificial construction of specific misorientations from single crystals
  - Harvesting of desired misorientation from large grained sample
- **Alternate laser-based characterization techniques may offer opportunities**

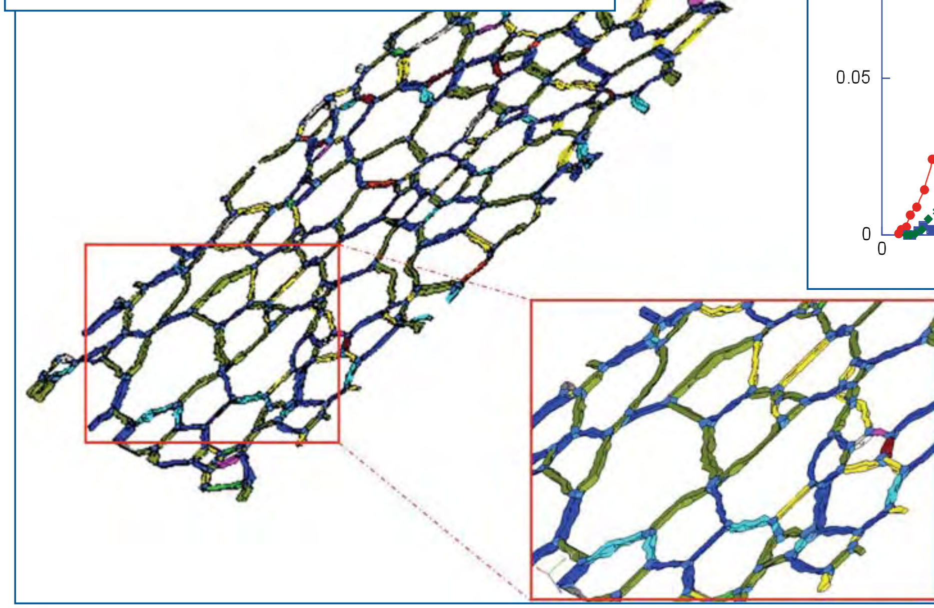
Measurement of Grain Boundary Effects: Path Forward

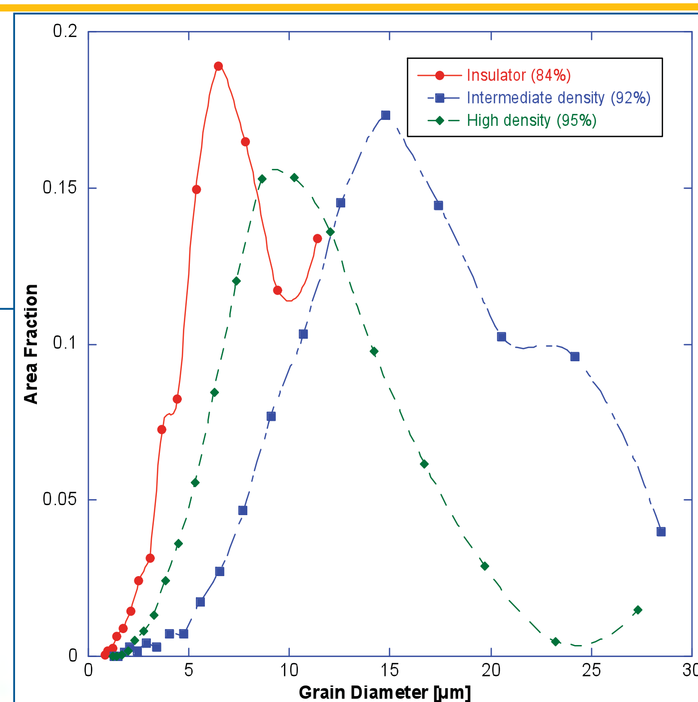
GRAIN BOUNDARIES K_{GB}

OIM of UO_2 pellet



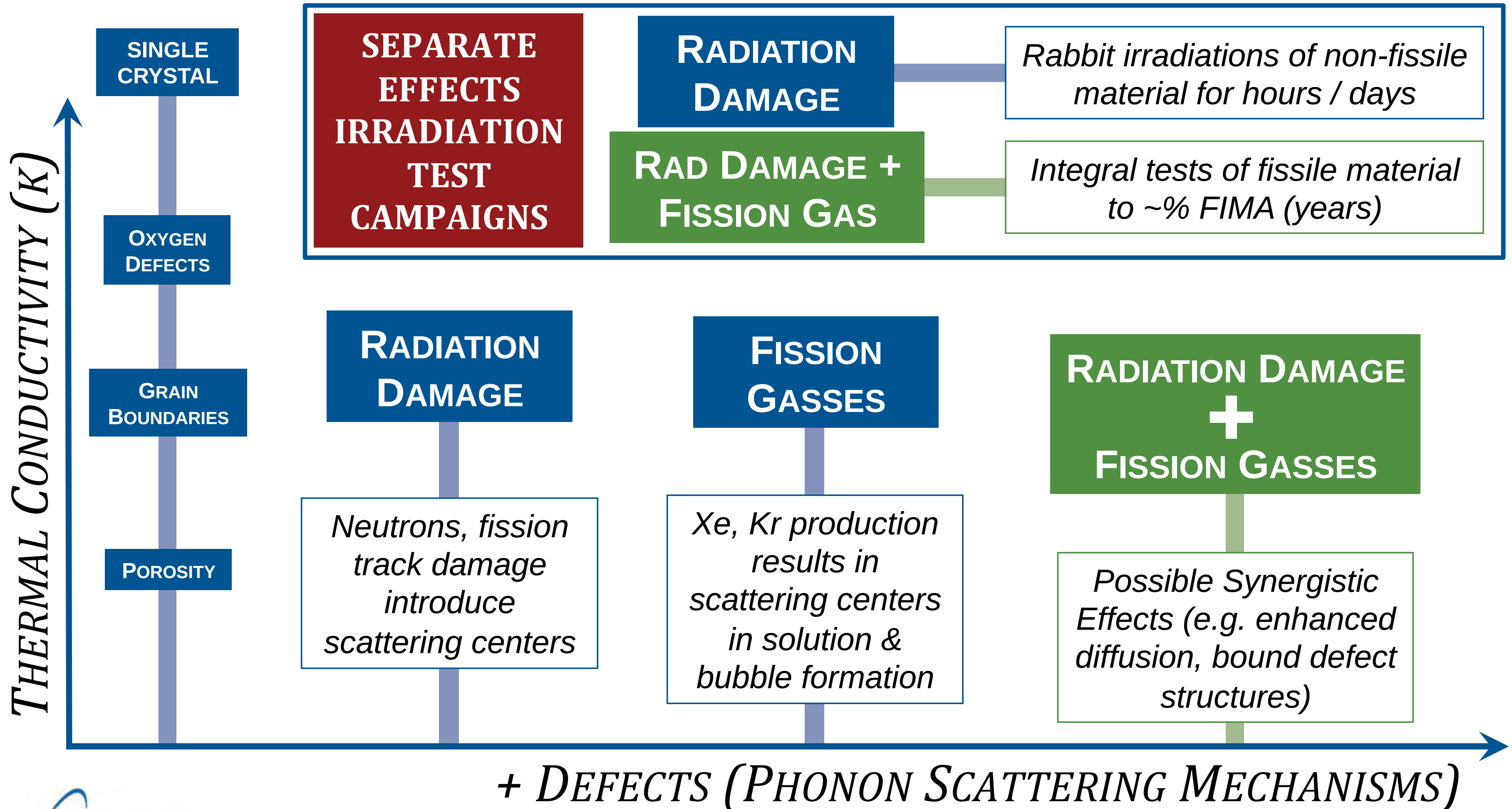
Map of boundary types from EBSD data





Statistical analysis of boundary type fraction for different UO_2 pellets

Sample Mechanisms Governing Thermal Transport in Irradiated Ceramic Systems



Ongoing and FY12 Work

- **Melt point studies for UO_2, $(U,La)O_2$, $(U,Ce)O_2$, $(U,Nd)O_2$ *Summer 2011***
- **Development of thermal conductivity curves for UO_2 as a function of O/M (0.02 intervals) in fluorite phase field to 1600°C *Summer 2011***
- **Texture / porosity effects studies on thermal transport *FY12***
- **Separate effects based irradiation campaigns (rabbit testing at HFIR) *FY12***

Sintering of mixed-oxide fuel pellets

J. Mitchell, M. Chavez (MST-16); S. Willson (MET-1); K. McClellan (MST-8)

We are measuring the thermophysical properties of mixed-oxide (MOX) and minor actinide-bearing MOX (MAMOX) fuel pellets in support of the Fuel Cycle Research and Development program (FCR&D). These properties include heat capacity, thermal expansion, density, and elastic constants. To better understand these properties and how processing impacts them, we have been studying the sintering behavior of fuel pellets as a function of temperature and atmosphere.

MOX and MAMOX pellets were sintered in a dilatometer in Ar or Ar-6% H_2 atmospheres to collect dynamic dimensional changes from room temperature to 1500 °C. Similar samples were subjected to interrupted sintering, where they were heated to 600, 800, 1000, 1200, or 1400 °C in a furnace for comparison of their pre- and post-sintering dimensions. The interrupted sintering samples have onset temperatures lower than those measured using the dilatometer, which may reflect a slight difference in the RT to 400 °C heating rates and subsequent differences in the oxygen:metal evolution.

Plutonium science and research strategy: Use of special isotopes – ^{242}Pu

D.L. Clark (INST-OFF); E.D. Bauer (MPA-CMMS); D.S. Peterson (MST-7)

Under the NNSA Complex Transformation, Los Alamos National Laboratory was named a consolidated Center of Excellence for plutonium research, development, and manufacturing activities. In response, the Laboratory has developed an institutional plutonium science strategy that defines research needs and opportunities that share a common underpinning of Los Alamos's missions in nuclear deterrence, global threat reduction, and energy security. One of our 14 objectives is to establish a small-scale plutonium research and development effort using plutonium-242, and other scarce and unique materials and isotopes.

Having a 16 times lower specific activity than Pu-239, meaningful quantities of Pu-242 metal, alloys and compounds can be used in radiological facilities that function under lower security and lower hazard levels than Category I, II, or III facilities. By focusing on unique, low-specific activity isotopes like Pu-242 the whole nature of Pu research can be dramatically improved, with reduced costs and the possibility of not having the specimens change during measurements. This provides the opportunity to engage academic collaborators and make better use of national user facilities for Pu research. It will mandate the ability to recover materials in a cost effective way. Success will demand a coordinated, integrated effort between the chemistry, physics, and materials science communities. New approaches (low-loss technologies) need to be developed and demonstrated to successfully recover and recycle these rare isotopes. An update on progress is presented.

Process modeling of plutonium and uranium casting

D.A. Korzekwa (MST-6); J.W. Gibbs (Northwestern U.); D.R. Korzekwa,F.J. Freibert (MST-16); Telluride project team (ASC)

Truchas is a multi-physics code being developed at Los Alamos for simulating materials processes such as metal casting and thermal processing. The capabilities are tailored specifically for vacuum induction casting of reactive metals such as uranium and plutonium alloys. Truchas has been funded by the Advanced Simulation and Computing program and is designed to run high-resolution simulations on massively parallel computing platforms.

Two examples of the use of process simulation to aid in the development and understanding of casting processes are presented. Collaborating with Idaho National Laboratory, we are modeling an experimental process for casting a metallic fuel composed primarily of uranium, zirconium and plutonium. The simulations provide insight into the fluid flow and heat transfer behavior to help evaluate and optimize the process. Another example describes the use of modeling to develop a mold design and induction heating schedule to closely control the solidification and cooling of alpha phase plutonium test samples. The first iteration of this design produced very high quality material and is a significant improvement over recent previous casting designs.

LEU U-10Mo research reactor fuel development and scale up

D. Dombrowski, R. Aikin, D. Alexander, A. Clarke, and K. Clarke (MST-6); T. Claytor (AET-6); J. Crapps, A. Duffield, P. Dunn, R. Edwards, R. Forsyth, D. Hammon, J. Katz, P. Kennedy, D. Korzekwa (MST-6); N. Mara (MPA-CINT); B. Mihaila, C. Necker, P. Papin, M. Pena, K. Rau, R. Schulze, V. Vargas, R. Weinberg (MST-6)

DOE NA-21 executes a global mission to convert HEU reactors to LEU fuel to reduce the worldwide inventory of weapons usable materials. The NA-21 CONVERT program is developing the supply of special high-density LEU U-10Mo fuel to support these research reactor conversions. This high visibility, schedule-driven project will convert U.S. High Performance Research Reactors (NBSR, MITR, ATR, MURR, HFIR) to LEU fuel; NRC reactors to be done by 2015. LANL will perform integrated process scale ups, develop alternative process steps and perform advanced characterization, especially bond strength. This poster shows highlights of LANL science and technology contributions that will enable NA-21 to qualify the fuel and provide it for reactor conversion.

Residual stresses in aluminum clad uranium-10wt%molybdenum fuel plates

D.W. Brown, L. Balogh (MST-8); B. Clausen (LANSCE-LC); M. Okuniewski (Idaho National Laboratory)

The mission of the Global Threat Reduction Initiative (GTRI) of the National Nuclear Security Administration (NNSA) in the U.S. Department of Energy is to reduce and protect vulnerable nuclear and radiological material located at civilian sites worldwide by providing support for the countries' own national programs. The GTRI Reactor Convert program converts research reactors from the use of highly enriched uranium (HEU) to low enriched uranium (LEU). The baseline fuel for conversion of high performance research reactors is aluminum clad monolithic uranium-10wt.% molybdenum (U10Mo). One bonding technique for the fuel to the cladding is hot isostatic pressing. The thermal expansion of U10Mo is roughly half that of aluminum, so a significant residual stress is expected following cooling from the pressing temperature. The residual stress field was measured with 0.1 mm resolution on mini fuel plates (0.25 mm thick U10Mo, 1 mm thick Al-clad) in transmission with high-energy x-ray diffraction at beamline
1ID-C at the Advanced Photon Source. In-plane compressive stresses approaching 250 MPa were observed in the U10Mo, suggesting balancing yield-level tensile residual stresses in the aluminum cladding.

Chemical segregation of U-10Mo fuel foils during simulated bonding cycles using neutron diffraction

S.C. Vogel (LANSCE-LC); D.W. Brown (MST-8); M. Okuniewski (Idaho National Laboratory)

We re-investigated the TTT diagram of U-10 wt.% Mo using isothermal high temperature neutron diffraction between 420°C and 560°C. U-10 wt.% Mo foils are fuel for the GTRI Reactor Convert program. Samples were ~200μm thick U-10Mo fuel foils with Zr cladding of 25μm on each side. Decomposition and ordering are competing processes, therefore, both α-U and an ordered γ-related phase γ^1 occur. Neutron diffraction uniquely allows identification of the involved phases and provides weight fractions as well as lattice parameters, crystal structures and textures as a function of holding time. From the lattice parameters, the Mo concentration in the residual γ-phase was estimated, showing enrichment in Mo. The texture showed a preference of the decomposition for specific crystal orientations, resulting in a substantial preferred orientation in both α-U and γ^1.

Neutron diffraction proved to be a valuable tool to obtain a complete picture of such processes, without the deficiencies of classical methods such as DSC (not phase sensitive), dilatometry (sensitive to length changes due to transformation as well as Mo re-partitioning), metallography or x-ray diffraction (surface sensitive).

Radioparagenesis: Robust nuclear waste form design and novel material discovery

B. Dorado (MST-8); C. Jiang (Central South University, China); K. Sickafus (MST-8); B. Scott (MPA-MC); M. Nortier, M. Fassbender, L. Wolfsberg (C-IIAC); B. Uberuaga, C. Stanek (MST-8)

Nearly 30 years ago, Gray [1] and Vance, Roy, et al. [2] posed the question: if a major constituent of a crystalline compound is an unstable isotope, what is the effect of the transmutation of that isotope to a chemically different element on the structure and stability of the compound? These researchers were explicitly interested in the impact of this transmutation on the performance of ceramic nuclear waste forms. However, since the half-lives of the so-called "short-lived" isotopes in the nuclear waste stream (e.g. ^{90}Sr and ^{137}Cs) is approximately 30 years, corresponding experiments require more than 100 years. As such, a systematic answer to the transmutation question has remained elusive.

We have recently revisited the transmutation question with modern computational materials science tools. Using density functional theory calculations, we have attempted to reproduce the evolution of a simplified example crystalline waste form, $^{137}CsCl$, during transmutation of ^{137}Cs to ^{137}Ba via β- decay, *i.e.* $^{137}Cs_{1-x}{}^{137}Ba_xCl$. By surveying the lattice energies of all possible crystal structures for a range of x, we predict that the rocksalt structure is thermodynamically favored (over the CsCl structure) at $x \approx 0.2$ [3], and this preference increases with increasing x. Based on this result (and similar results for other systems), we have introduced the concept of radioparagenesis, which we define as the formation of compounds, often unconventional (e.g. rocksalt BaCl), due to the chemical transmutation that occurs during in situ radioactive decay.

In this presentation, implications of radioparagenesis on nuclear waste form design [4], unconventional defect chemistry [5] and novel materials discovery [6] will be discussed. Also, recently devised experiments aimed at confirming radioparagenesis by significantly accelerating the transmutation process, using small quantities of highly radioactive samples, will be discussed. These experiments require a compromise between sample size and sample activity – that is, enough sample must be produced (typically via an accelerator-based process) that it can be characterized, but the amount must be kept small in order to allow for safe handling. Furthermore, choice of isotope (chemically distinct daughter, half-life conducive to experimentation, etc) is an important consideration. Results of these experiments will be compared to DFT calculations.

[1] W.J. Gray, "Fission product effects on high-level radioactive waste forms," *Nature*, **296** (1982) 547-549.
[2] E.R. Vance, R. Roy, J.G. Pepin and D.K. Agrawal, "Chemical mitigation of the transmutation problem in crystalline nuclear waste radiophases," *Journal of Materials Science*, **17** (1982) 947-952.
[3] C. Jiang, C.R. Stanek, N.A. Marks, K.E. Sickafus and B.P. Uberuaga, "Predicting from first principles the chemical evolution of crystalline compounds due to radioactive decay: The case of the transformation of CsCl to BaCl," *Phys. Rev. B*, **79** (2009) 132110.
[4] C. Jiang, B.P. Uberuaga, K.E. Sickafus, F.M. Nortier, J.J. Kitten, N.A. Marks, and C.R. Stanek, "Using 'radioparagenesis' to design robust nuclear waste forms," *Energy and Environmental Science*, **3** (2010) 130.
[5] B.P. Uberuaga, C. Jiang, C.R. Stanek, K.E. Sickafus, N.A. Marks, D.J. Carter and A.L. Rohl, "Implications of transmutation on the defect chemistry in crystalline waste forms," *Nucl. Inst. Meth. B*, **268** (2010) 3261.
[6] C. Jiang, C.R. Stanek, N.A. Marks, K.E. Sickafus, B.P. Uberuaga, "Radioparagenesis: The formation of novel compounds and crystalline structures via radioactive decay," *Phil. Mag. Lett.* **90** (2010) 435.

Fissionable scintillators for measuring neutron flux

S. Stange (N-2); E.I. Esch (N-1); R.E. Del Sesto (MPA-MC); R.E. Muenchausen (MST-7); F.L. Taw (C-IIAC); F. Tovesson (LANSCE-NS); E.A. Burgett (Idaho State University)

Fission chambers and ionization chambers containing one or more foils thinly coated with a fissile or fissionable material are commonly used to measure neutron flux for nuclear physics experiments such as those performed at the Los Alamos Neutron Science Center (LANSCE). These detectors are easy to operate, are not damaged by radiation, and have an acceptable efficiency for many applications. However, they also are prone to pulse pile-up, require long measurement times, and are bulky and fragile.

We proposed to address these issues by developing a nanocomposite fission detector, consisting of nanoparticles of fissionable material dispersed in a scintillating matrix. This would produce a detector that can be loaded with up to two orders of magnitude more uranium and has a pulse rise time that is two orders of magnitude faster. In addition, the use of a plastic matrix would make the detector far more robust than current fission chambers.

Material attractiveness in nuclear fuel cycles

C. Bathke (D-5)

We must anticipate that the day is approaching when details of nuclear weapons design and fabrication will become common knowledge. On that day we must be particularly certain that all special nuclear materials are adequately accounted for and protected. Among other things this requires a clear understanding of the utility of nuclear materials to potential adversaries. To this end, this poster examines the attractiveness of materials mixtures containing special nuclear materials (SNM) and alternate nuclear materials (ANM) associated with the PUREX, UREX, COEX, and THOREX reprocessing schemes. This paper provides a set of figures of merit (FOM) for evaluating material attractiveness that covers a broad range of proliferant state and subnational group capabilities. The primary conclusion of this paper is that all fissile material needs to be rigorously safeguarded to detect diversion by a state and provided the highest levels of physical protection to prevent theft by subnational groups; no “silver bullet” fuel cycle has been found that will permit the relaxation of current international safeguards or national physical security protection levels. The work reported herein has been performed at the request of the United States Department of Energy (DOE) and is based on the calculation of "attractiveness levels" that are expressed in terms consistent with, but normally reserved for nuclear materials in DOE nuclear facilities. The methodology and findings are presented. Additionally, how these attractiveness levels relate to proliferation resistance and physical security are discussed.

Materials for clean energy applications

D.E. Watkins (MPA-DO)

Materials for clean energy applications is a $40M activity spanning use-inspired basic research to the development and testing of prototype energy technology systems. LANL's key technical areas can be broken out as materials for energy conversion, storage, and transmission. I will describe technical highlights and discuss future directions for LANL contributions to America's energy security.

Overview:
Materials for Clean Energy Applications

David Watkins

MPA Deputy Division Leader

2 June 2011

UNCLASSIFIED

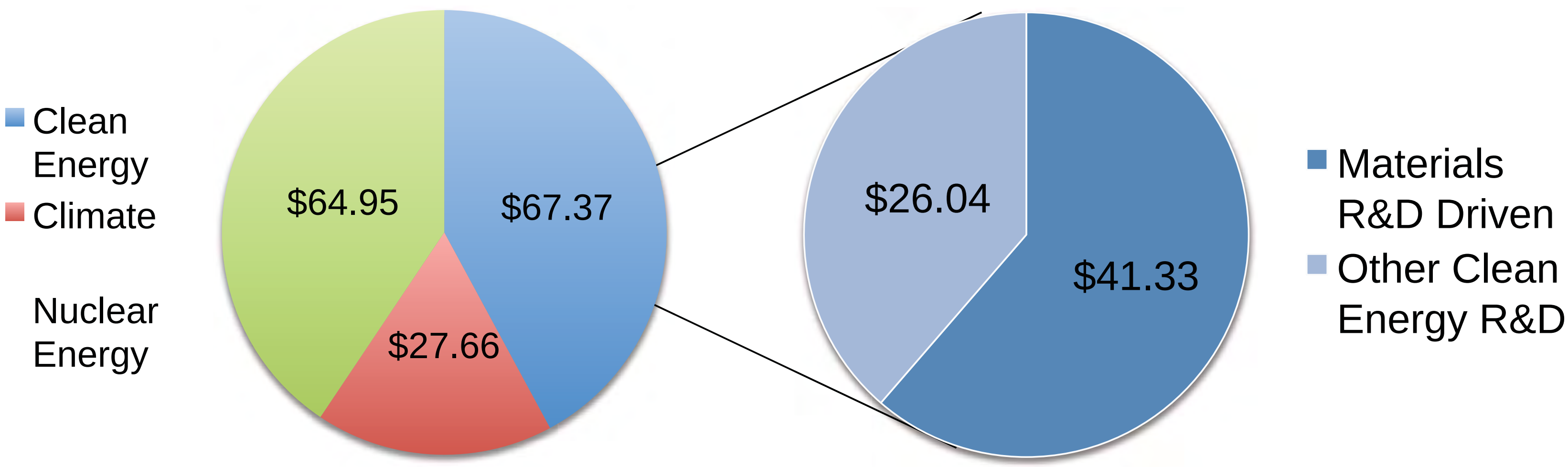
Emergent Phenomena: Thrust areas further define the 'Areas of Leadership' for the Materials Pillar

- **Integrated Nanomaterials**
  - Reduced dimensionality materials for control of emergent functionality
  - Center for Nanophotonics
- **Complex Functional Materials**
  - Functional materials for energy conversion, storage, and transmission
  - Materials inspired by living systems
  - Multifunctional adaptive materials
- **Materials in Radiation Extremes**
  - Advanced radiation & temperature tolerant structural materials
  - Advanced nuclear fuels & nuclear waste materials
- **Actinides and Correlated Electron Materials**
  - Understanding and controlling emergent electronic states
  - Actinide materials science center of excellence
  - Predicting and controlling plutonium aging and lifetime
- **Materials Dynamics**
  - Linking microstructure to macroscopic behavior under dynamic loading
  - Observation-to-control of dynamic processes
  - Next generation diagnostics and drivers
- **Energetic Materials**
  - Prediction and control of safety, initiation and performance of explosives

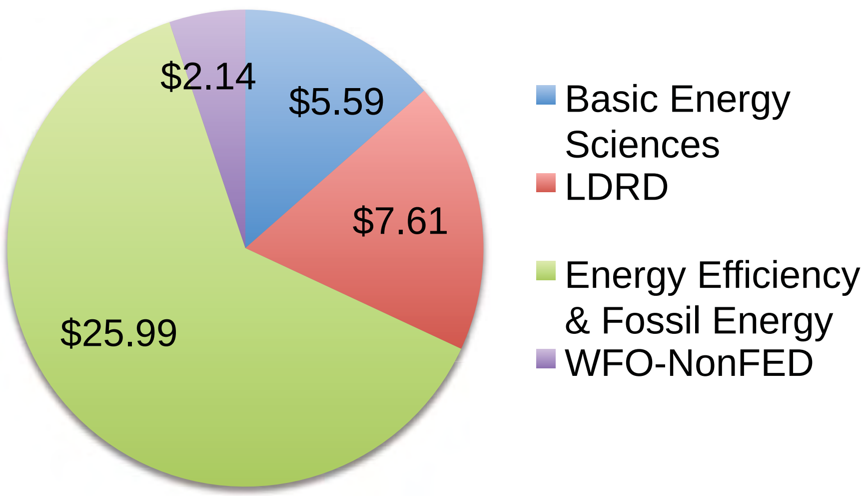
Clean and Renewable Energy is one of three thrusts in LANL's Energy Strategy

FY-10 Energy Projects at LANL (M$)

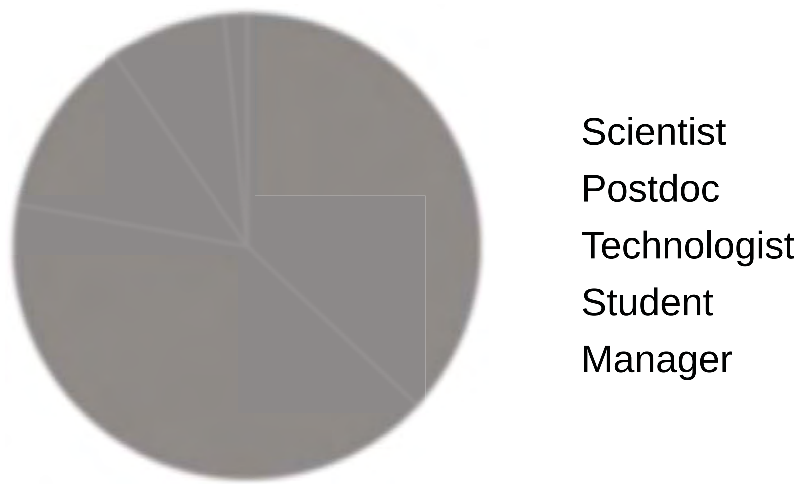
FY-10 Clean Energy



LDRD and DOE applied energy programs dominate as funding sponsors

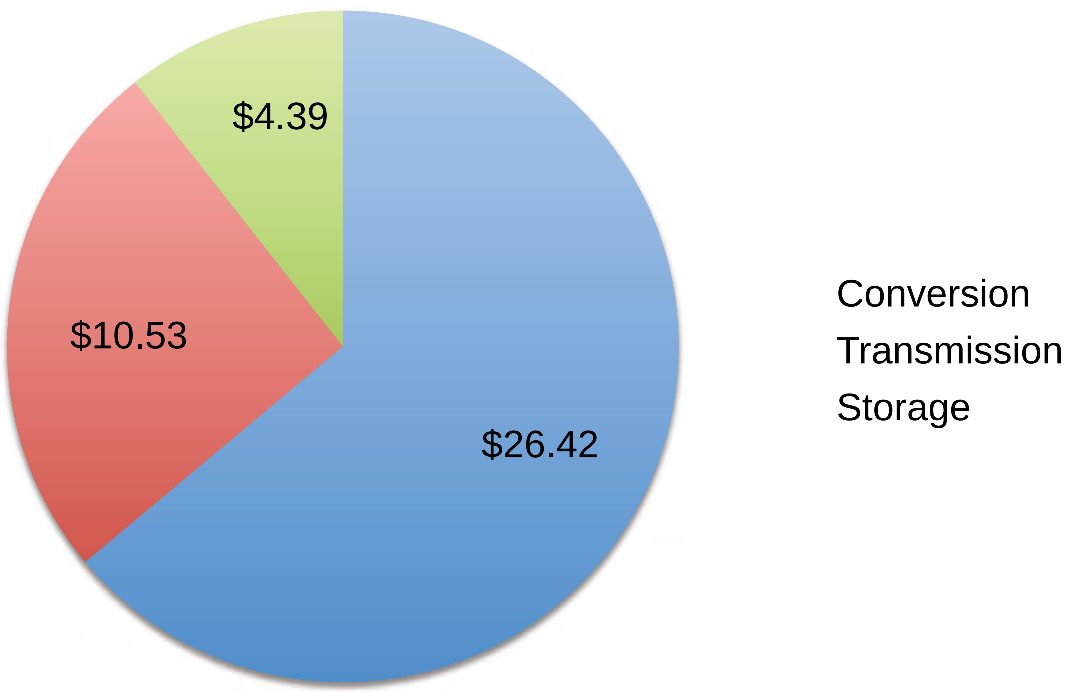


Funding Source in M$



~120 FTEs are devoted to Materials for Clean Energy R&D

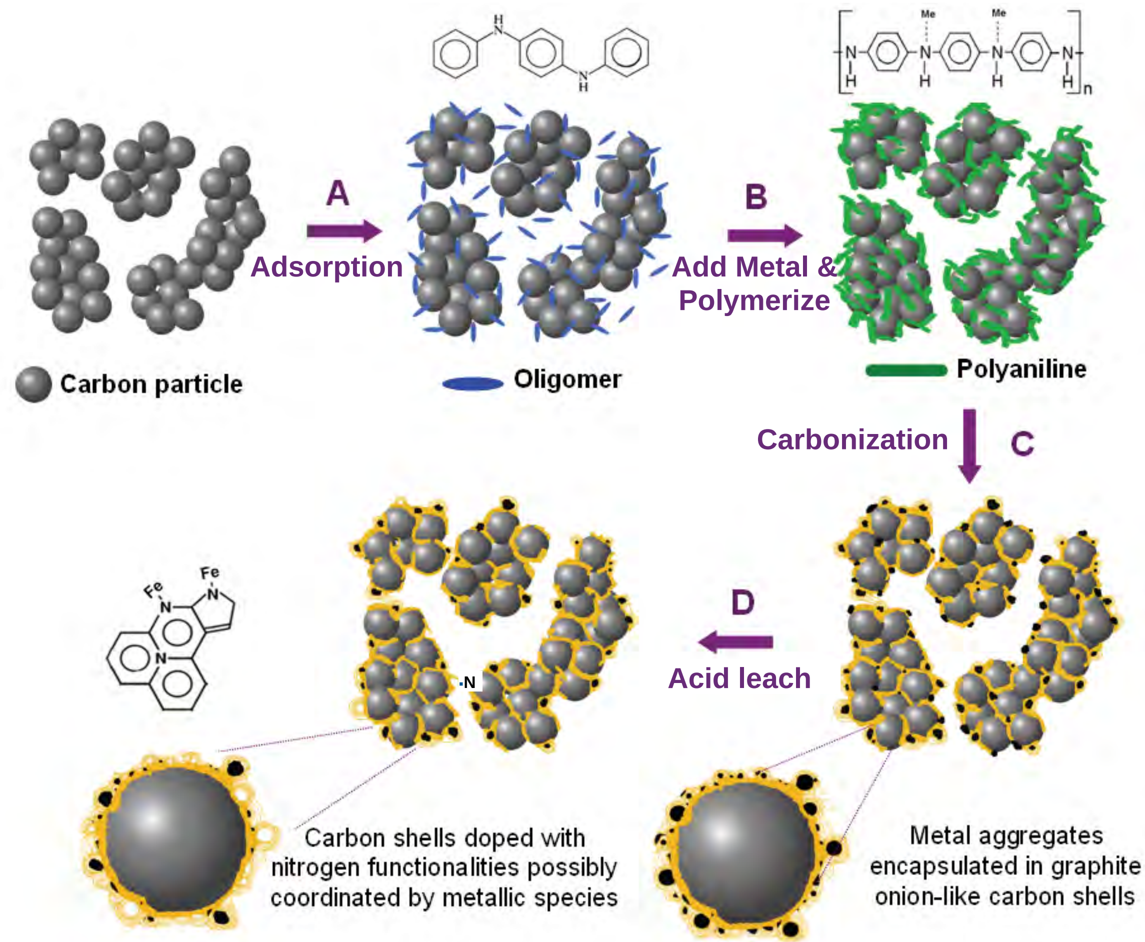
Materials for clean energy divides into three themes

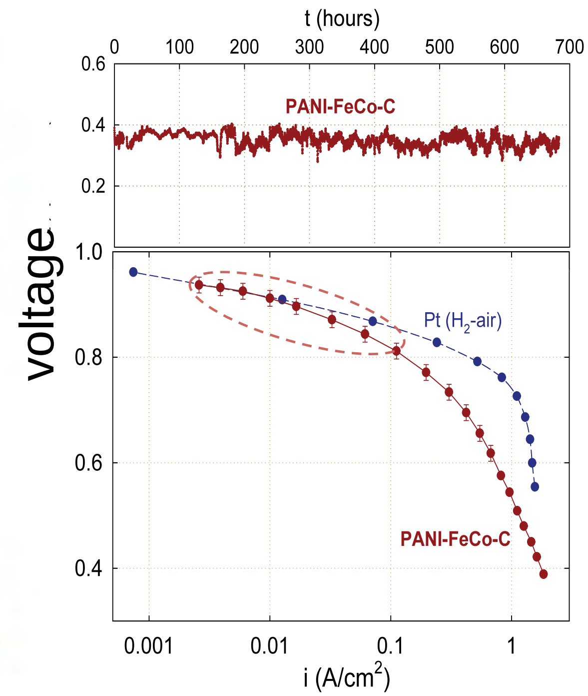


Materials-driven clean energy research

- **Conversion**
  - Photophysics, photovoltaics, and solid state lighting
  - Fuel Cells
  - Fossil energy and chemical separations
- **Transmission**
  - Superconducting power transmission
- **Storage**
  - Hydrogen storage
  - Grid-scale storage

Energy conversion: First stable, high-performance non-precious metal fuel cell cathode

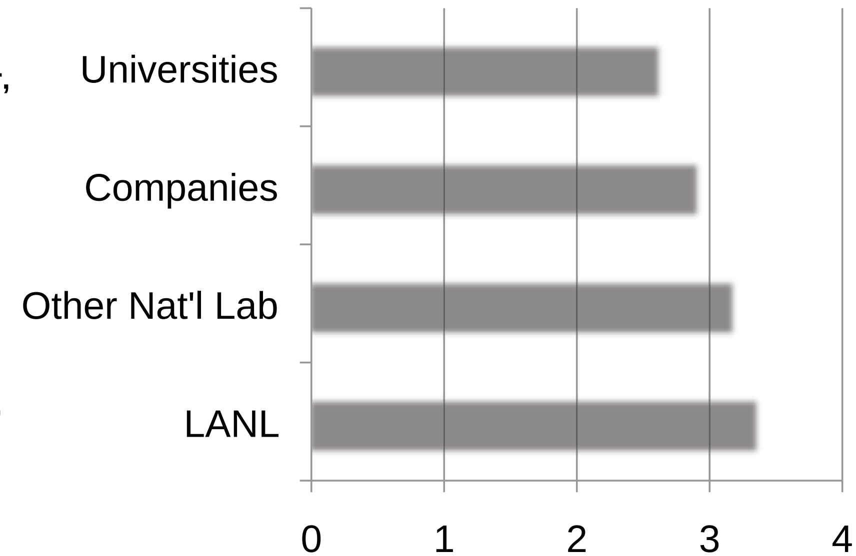




High-Performance Electrocatalysts for Oxygen Reduction Derived from Polyaniline, Iron, and Cobalt, G. Wu, K. L. More, C. M. Johnston, and P. Zelenay, *Science*, 332, 6028, 443-447 (2011)

The impact of LANL's fuel cell program derives from use-inspired research

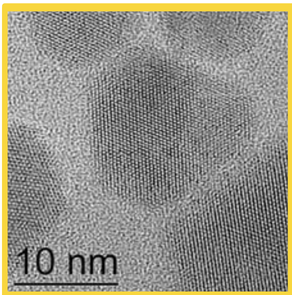
- **Scientific aspects of polymer electrolyte fuel cell durability and degradation, Borup, R; Meyers, J; Pivovar, B, et al. CHEMICAL REVIEWS 107, 10, 3904-3951, (2007)**
  - Times Cited: 281
- **A class of non-precious metal composite catalysts for fuel cells Bashyam, R; Zelenay, P, NATURE, 443, 7107, 63-66 (2006)**
  - Times Cited: 263
- **Electrocatalysis in direct methanol fuel cells: in-situ probing of PtRu anode catalyst surfaces, Dinh, HN; Ren, XM; Garzon, FH, et al. JOURNAL OF ELECTROANALYTICAL CHEMISTRY 491, 1, 222-233 (2000)**
  - Times Cited: 155
- **Direct methanol fuel cells: progress in cell performance and cathode research Thomas, SC; Ren, XM; Gottesfeld, S, et al., ELECTROCHIMICA ACTA 47, 22, 3741-3748, (2002)**
  - Times Cited: 136



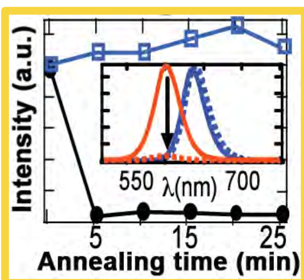
LANL leads in average score through DOE peer review of ongoing projects and total projects awarded through competitive processes.

Energy conversion: Quantum dots

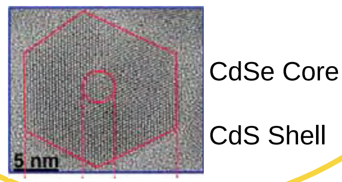
Epitaxial-quality" QDs grown in a flask



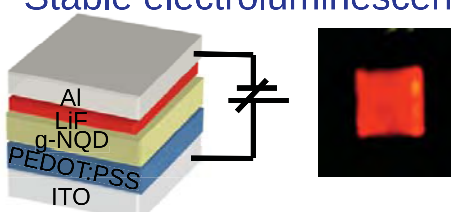
Robust to chemical & thermal treatments



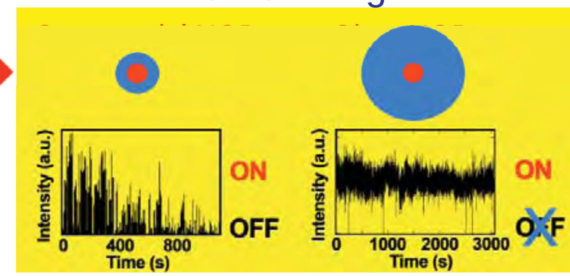
Novel physical & electronic nanostructure affords new functional class of QD for device (LED, single-photon, lasing) and tracking applications



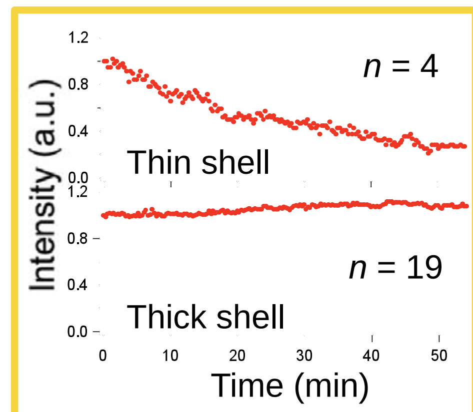
Stable electroluminescence



Non-blinking



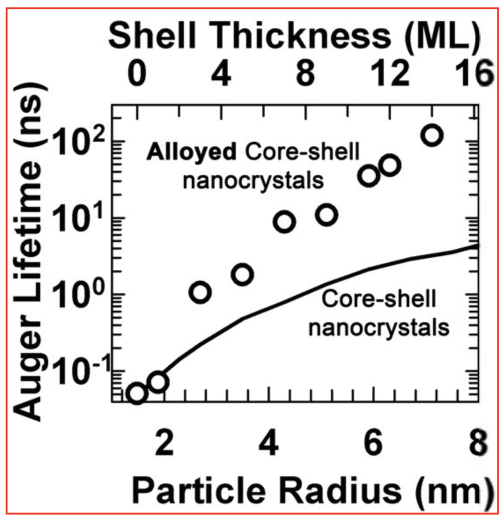
Non-photobleaching



Chen et al. *J. Am. Chem. Soc.* (2008) 5026 (99 citations)

New research theme: *Interfacial Alloying for "Auger Recombination Engineering"*

- **Vela et al. *J. Biophotonics* (2010), 706**
- **Hollingsworth et al. U.S. Patent 7,935,419**
- **GarcÍa-SantamarÍa et al. *Nano Lett.* (2009), 3482**
- **Htoon et al. *Nano Lett.* (2010), 2401**
- **S. Brovelli et al. Nature Communications (2011) 1281**



C&EN
CHEMICAL & ENGINEERING NEWS

January 17, 2011 Volume 89, Number 3 p. 32

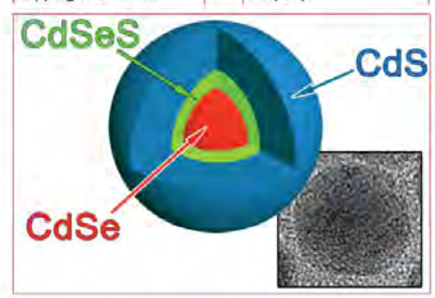
Interfacial Properties Boost Lasing by Quantum Dots

Two-monolayer-thin alloy layer in core-shell nanocrystals aids optics applications

Mitch Jacoby

Quantum dots' prospects for use in laser applications are looking bright as a result of a study showing how to limit a deleterious effect that robs the semiconductor nanocrystals of their potential lasing power (*Nano Lett.*, DOI: 10.1021/nl103801e). Ten years ago, a research team that included Victor I. Klimov of Los Alamos National Laboratory showed that quantum dots could be made to lase, a demonstration that opened the door to several applications in optics.

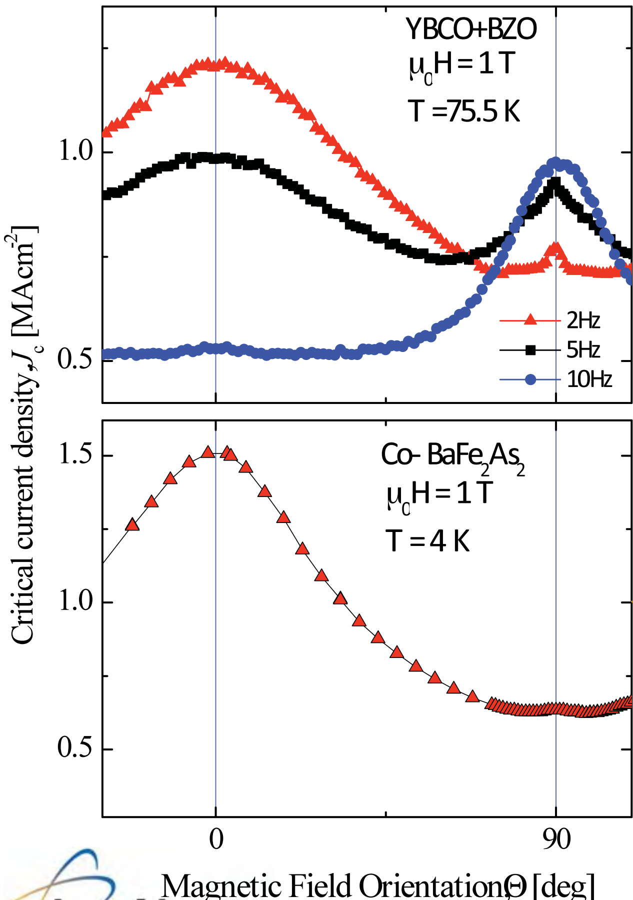
Despite the proof-of-principle experiment, nanocrystal lasing has remained impractical because of a fast relaxation process known as Auger recombination, which quenches the electronic excitations required for lasing and causes electron, rather than photon, emission. Now, Klimov, Florencio García-Santamaría, Sergio Brovelli, and coworkers report that capping a cadmium selenide core with just a few monolayers of cadmium sulfide suppresses the Auger process by more than two orders of magnitude. The group's spectroscopic measurements indicate that the improvement stems from the unique electronic properties of a two-monolayer-thin alloy layer at the core-shell interface, a finding consistent with recent theoretical predictions, they say.



Schematic and transmission electron micrograph of a quantum dot nanocrystal with a cadmium selenide core and a cadmium sulfide shell separated by a thin alloyed interface formed due to intermixing between selenium and sulfur atoms.

Los Alamos
NATIONAL LABORATORY
EST. 1943

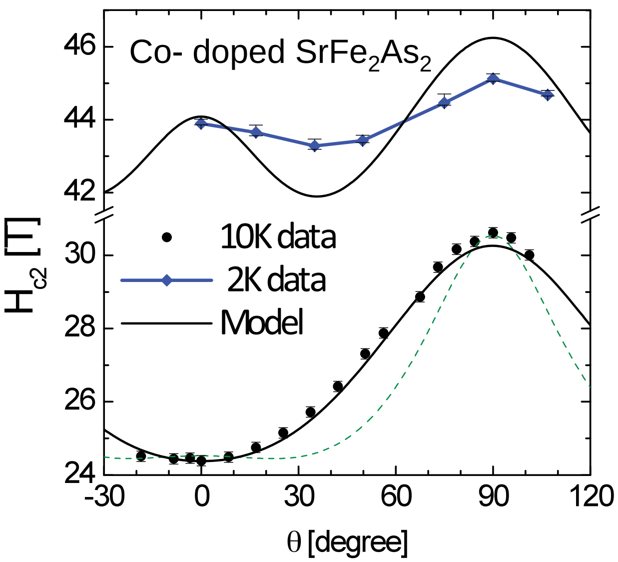
Oxide- and iron-based superconductor pinning landscape



New Materials

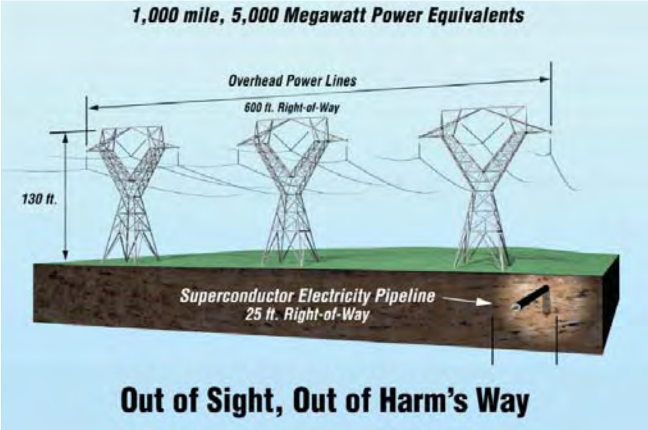
Technologically relevant: Engineering of pinning landscape leads to world-record critical currents

Scientifically relevant: Additional complexity due to multi-band



B. Maiorov *et al* Nat. Mat. **8** (2009), B. Maiorov *et al* SuST **24** (2011)

LANL has contributed to transmission test beds and is developing new concepts



AMSC transmission level cable

- First in US grid, 138 kV, 574 MVA
- Operator: Long Island Power Authority
- The three cables shown entering the ground can carry as much power as all of the overhead lines on the far left.
- In service since 4/22/08

Upgrade Transmission Line

- 5x the power, same voltage
- Same Right of Way, No new permits required, No legal fee, No public meetings
- New cryostat and suspension concepts
- New cooling system
  - Longer distance between refrigeration stations
  - Low capital and running cost
  - High reliability
- New termination design

Highlights of applied superconductivity R&D

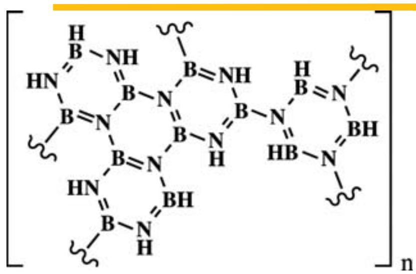
- **Initial development of YBCO based Coated Conductors**
  - X.D. Wu, S.R. Foltyn, P. Arendt, et al. Appl. Phys. Lett. 65, 1961 (1994) Times Cited: 170
  - X.D. Wu, S.R. Foltyn, P. Arendt, et al. Appl. Phys. Lett. 67, 2397 (1995) Times Cited: 280
- **Understanding vortex pinning in Coated Conductors**
  - L. Civale, B. Maiorov, A. Serquis, et al. Appl. Phys. Lett. 84, 2121 (2004) Times Cited: 124
- **Optimizing the vortex pinning landscape in Coated Conductors**
  - J.L. MacManus-Driscoll, S.R. Foltyn, Q.X. Jia, et al. Nature Materials 3, 439 (2004) Times Cited: 376
  - S.R. Foltyn, H. Wang, L. Civale, et al. Appl. Phys. Lett. 87, 162505 (2005) Times Cited: 79
  - S.R. Foltyn, L. Civale, J.L. MacManus-Driscoll, et al. Nature Materials 6, 631 (2007) Times Cited: 124
  - T. G. Holesinger, L. Civale, B. Maiorov, et al. Advanced Materials 20, 391 (2008) Times Cited: 32
  - B. Maiorov, S.A. Baily, H. Zhou, et al. Nature Materials 8, 398 (2009) Times Cited: 46
  - M. Miura, S.A. Baily, B. Maiorov, et al. Appl. Phys. Lett. 96, 072506 (2010)
- **Applying the expertise and capabilities to the new iron-based superconductors**
  - S.A. Baily, Y. Kohama, H. Hosono, et al. Phys. Rev. Lett. 102, 117004 (2009) Times Cited: 33
  - B. Maiorov, T. Katase, S.A. Baily, et al. Supercond. Sci. Technol. 24, 055007 (2011)

LANL industrial & utility collaborations have been critical to technology impact

Previous (>30)

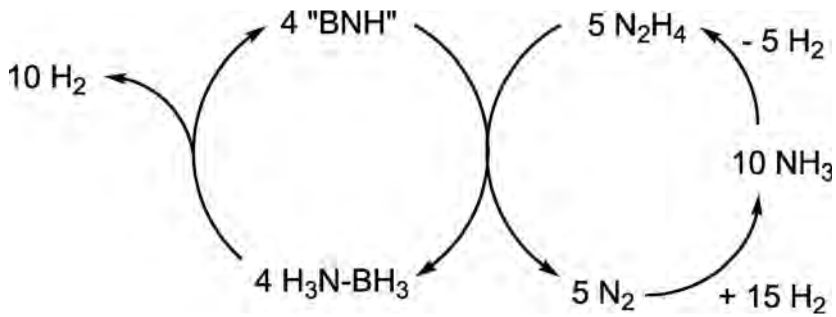
Present (8)

Energy storage: Advances in rechargeable solid hydrogen fuel storage

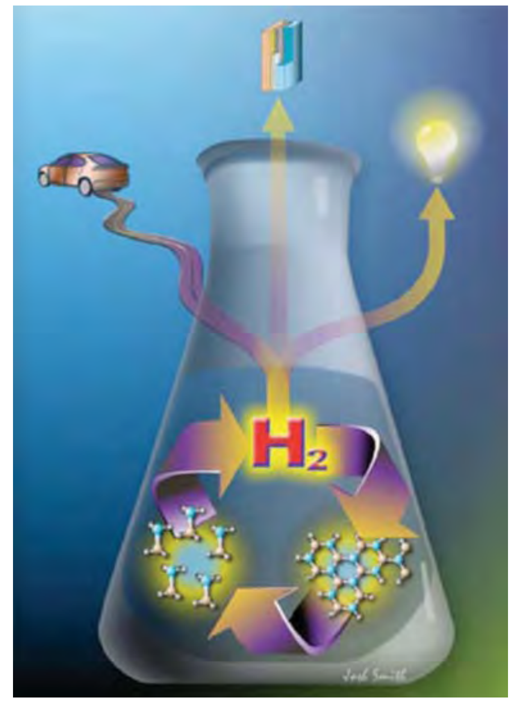


A representative structure of polyborazylene

Ideal overall reaction scheme for AB (H3N-BH3) regeneration from PB ("BNH") with hydrazine (N2H4).



- **New single-stage method for recharging the hydrogen storage compound ammonia borane**
- **Regeneration takes place in a sealed pressure vessel using hydrazine and liquid ammonia at 40°C**



"Regeneration of Ammonia Borane Spent Fuel by Direct Reaction with Hydrazine and Liquid Ammonia", ***Science*** *331, 1426 (2011); doi: 10.1126/science.1199003*

Future directions:
Key science and technology questions

- **What are the design principles for chemical functionality of materials?**
  - How do we control nanoscale properties to achieve macroscale functionality for energy applications?
  - What are the design principles that enable control of nanoscale structure and function? How can we exploit novel emergent phenomena for energy applications?
  - How can we design catalysts that enable the coupling of electron transfer and proton transfer to harvest the energy of diverse sources of hydrogen?
  - Can we develop modeling techniques for durability, efficiency, and cost-effectiveness of electrochemical systems?
- **What are the design principles that lead to new electronic/superconducting/magnetic materials?**
  - What is beyond Fermi liquid theory as a framework for describing interacting electrons?
  - What is the interplay between structure and correlations, and what are the consequences therein?
  - Can we understand why and how large magnetic moments form in 3d-electron metals?
  - Can we adequately control the microstructure of superconductors to optimize flux pinning across a broad range of applied field and materials?
  - Can we optimize HTS materials and fabrication methods to achieve commercial viability?

Key materials science capabilities

- Energy storage in chemical bonds
- Catalysis and phase transformations at surfaces and interfaces
- Materials structure-properties relations
- Functional materials for separations
- Diagnostics: from materials characterization and processing to prototype testing
- Sensor development
- Multi-scale modeling – from molecules to energy technology
- Integration – from basic science to engineered application

A flexible fabrication facility is key to expanding our capabilities and to future program development in Materials for Clean Energy

Presentations and posters

- **Victor Klimov:** Center for Advanced Solar Photophysics (EFRC)
- **Rod Borup:** Materials Development in the LANL Fuel Cell Program
- **Posters**
  - Anshu Pandey (C-PCS): Colloidal Materials for Light Harvesting Applications
  - John Stewart (C-PCS): A Comparative Study of Carrier Multiplication in PbSe and PbS Nanocrystals
  - Shadi Dayeh (MPA-CINT): Assessment of Silicon Nanowire Architecture for PV Application
  - Cynthia Welch (MST-7): Controlling Electrode Morphology to Improve Fuel Cell Performance and Durability
  - Eric Brosha (MPA-11): Nanoscale Ceramic Supports for PEM Fuel Cells
  - Katherine Berchtold (MPA-MC): Membrane Materials for Energy Applications
  - Ben Davis (MPA-MC): Chemical H_2 Storage Research at LANL
  - Leonardo Civale (MPA-STC): Comparative studies of vortex physics in oxide, iron-arsenide and MgB2 superconductors
  - Terry Holesinger (MPA-STC): Advances in HTS applications

Center for Advanced Solar Photophysics

V.I. Klimov (C-PCS)

This presentation provides a brief overview of research activities in the Center for Advanced Solar Photophysics with focus on spectroscopy of processes in semiconductor nanocrystals (NCs) of relevance to solar energy conversion. One such process is carrier multiplication (CM), or multiexciton generation, which can increase photocurrent in solar cells. Recent efforts in this area include: the development of fast, reliable screening methods for CM yields using photon counting with superconducting nanowire detectors; studies of the impact of “extraneous” processes on CM measurements; and the evaluation of the effects of the NC composition, size and shape on CM yields. As part of our effort on controlling excited-state dynamics, we study hot-electron transfer in NCs. We find that the efficiency of this process can approach 10% with incidental impurity-like acceptors, suggesting that even higher probabilities are possible with appropriately engineered acceptors. We also spectroscopically probe charge transport in NC-based exploratory devices such as optical field-effect transistors, unraveling the nature of conducting states in dark and under illumination, and helping to rationalize previously observed trends in carrier mobility and in the offset between photovoltage and nominal NC band gap. These studies illustrate how key insights into the performance of nanoscale materials are gained through close integration of spectroscopic, materials and device efforts across the center.

Center for Advanced Solar Photophysics:
Energy Frontier Research Center

Victor Klimov

C-PCS

Center for Advanced Solar Photophysics:
Energy Frontier Research Center

Center Participants

LANL: V. I. Klimov (Director),
J. M. Pietryga, I. Robel, H. Htoon, S. Tretiak

NREL: A. J. Nozik (Associate Director), M. C. Beard, J. C. Johnson, J. M. Luther, N. R. Neale

Rice University: N. J. Halas, P. J. Nordlander

UC Irvine: J. C. Hemminger, M. Law

University of Minnesota: U. R. Kortshagen

University of Colorado: D. M. Jonas

Colorado School of Mines: P. C. Taylor

George Mason University/NRL: Al. L. Efros

Center Focus: *Exploration of unique physics of nanoscale materials as a potential enabler of Generation III solar energy conversion technologies – the technologies that combine high efficiency with low fabrication cost.*

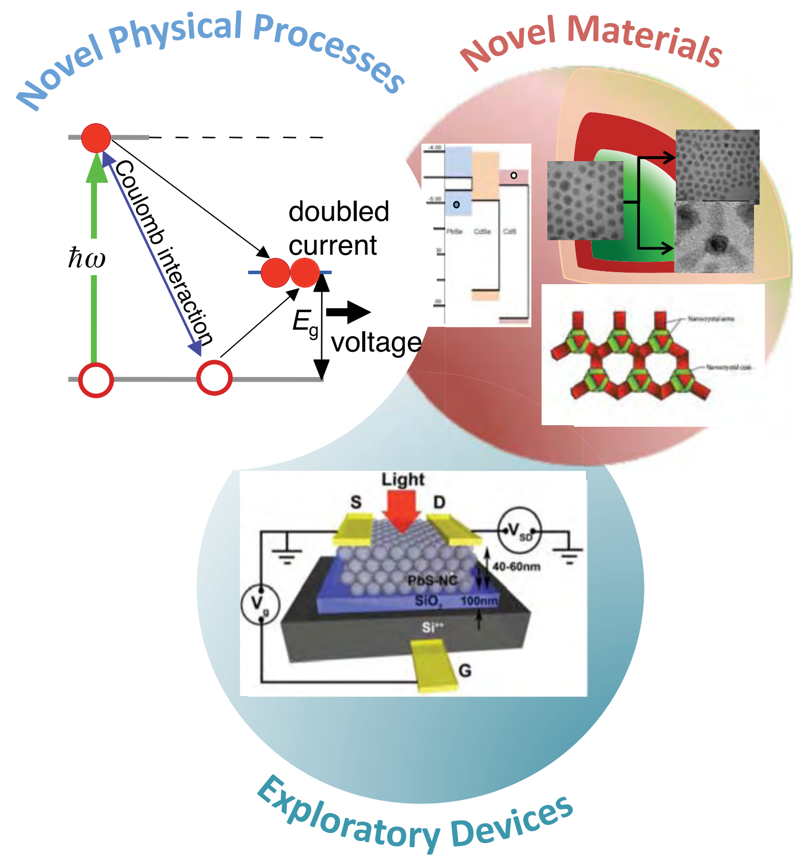
http://casp.lanl.gov

Novel physics, exploratory devices and engineered nanomaterials

- ***Thrust 1. Novel physical principles*** *for efficient capture and conversion of light into electrical charges*
  V. Klimov (LANL) & M. Beard (NREL)

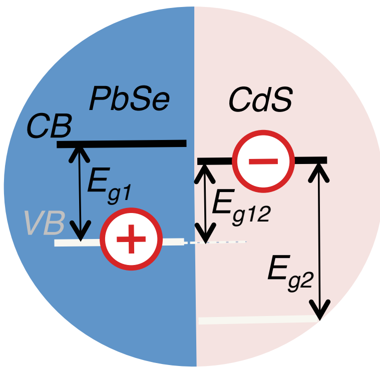
- ***Thrust 2. Charge manipulation on the nanoscale and exploratory device structures*** *that exploit the emergent physics of the nanosize regime*
  A. Nozik (NREL) & M. Law (UC Irvine)

- ***Thrust 3. Engineered nanomaterials*** *for probing and controlling the fundamental physics of the nanoscale size regime*
  J. Pietryga (LANL) & U. Kortshagen (U. Minnesota)

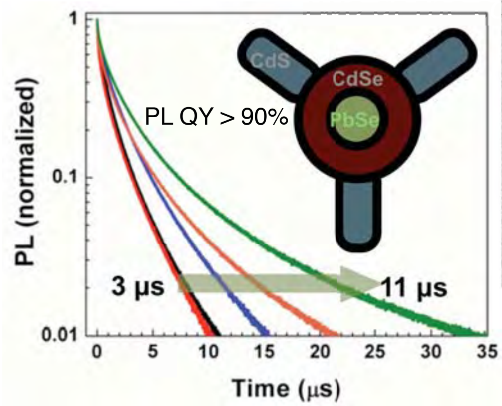


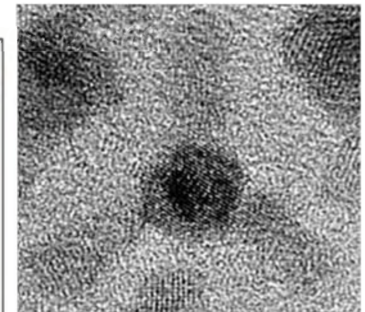
Novel materials: *Nanoscale junctions and semiconductor-metal hybrids*

- IR active Type-II Nanojunctions



- ✓ *Indirect gap (E_{g12}) tunable through IR*
- ✓ *Highly efficient charge separation*
- ✓ *Increased carrier lifetimes*
- ✓ *Potential applications in photocatalysis and PV*

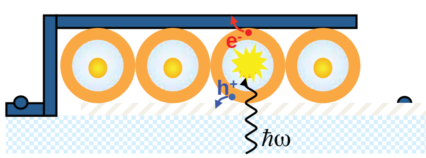


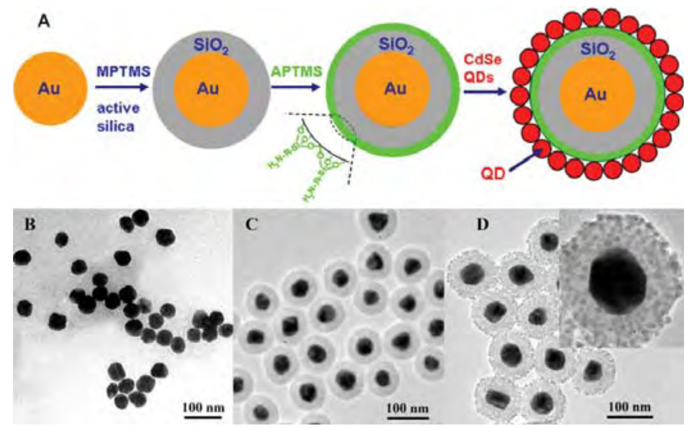


D. Lee et al. *J. Am. Chem. Soc.* **132**, 9960 (2010)

- Semiconductor-metal hybrids

- ✓ *Increased absorption cross sections*
- ✓ *Long-range energy transfer*
- ✓ *Potential applications in ultrathin solar cells*

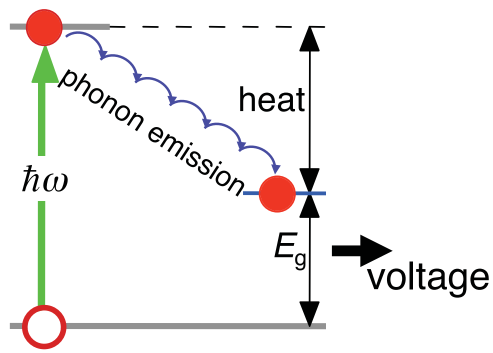


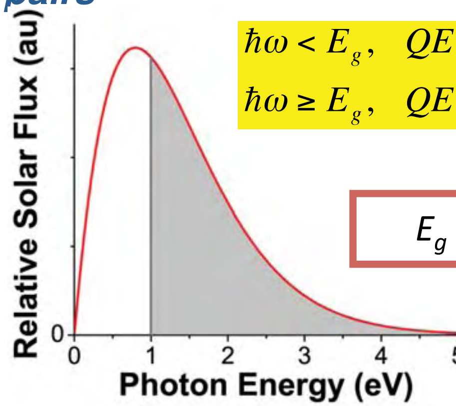


Novel physics: *Harvesting hot-electron energy via carrier multiplication*

- ***Traditional mechanism: 1 photon → 1 e-h pairs***

$\hbar\omega \geq E_g$





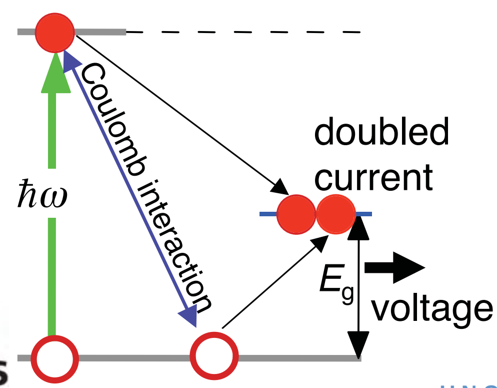
$\hbar\omega < E_g, \quad QE = 0$

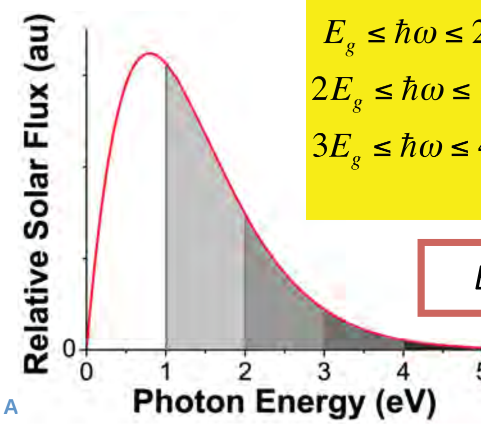
$\hbar\omega \geq E_g, \quad QE = 1$

E_g = 1.1 eV, η = **31%**

- ***Carrier multiplication mechanism*: *1 photon → 2 e-h pairs***

$\hbar\omega \geq 2E_g$





$\hbar\omega < E_g, \quad QE = 0$

$E_g \leq \hbar\omega \leq 2E_g, \quad QE = 1$

$2E_g \leq \hbar\omega \leq 3E_g, \quad QE = 2$

$3E_g \leq \hbar\omega \leq 4E_g, \quad QE = 3$

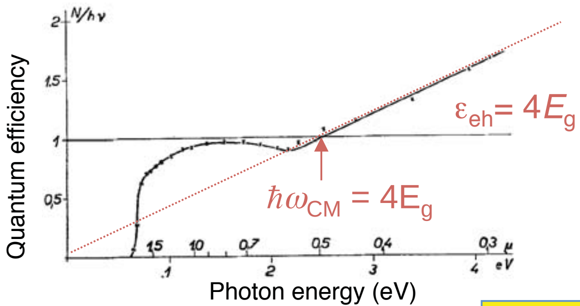
...

E_g = 0.45 eV, η = **42%**

Carrier multiplication in bulk semiconductors: *Some history*

- **Impact ionization in Ge (1957)**

S. Koc, *Czech. J. Phys.* **7**, 91 (1957)



ε_{eh} - *electron-hole pair creation energy*

$\hbar\omega_{CM}$ - *carrier multiplication threshold*

- **General trends in the bulk**

Energy & momentum conservation, phonon losses:

$\varepsilon_{eh} = E_g + \langle E_K \rangle + \langle E_{ph} \rangle$

$\langle E_K \rangle = 1.8\ E_g$

$\langle E_{ph} \rangle = 10 - 20\ E_L$

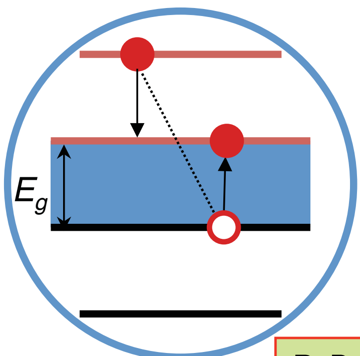
E-h pair creation energy (3E_g rule):

$\varepsilon_{eh} \geq 3E_g$

CM threshold:

$\hbar\omega_{CM} \geq 4E_g$

- **Semiconductor NCs**



Momentum conservation relaxation:

Can reduce $\langle E_K \rangle$

"Phonon bottleneck"

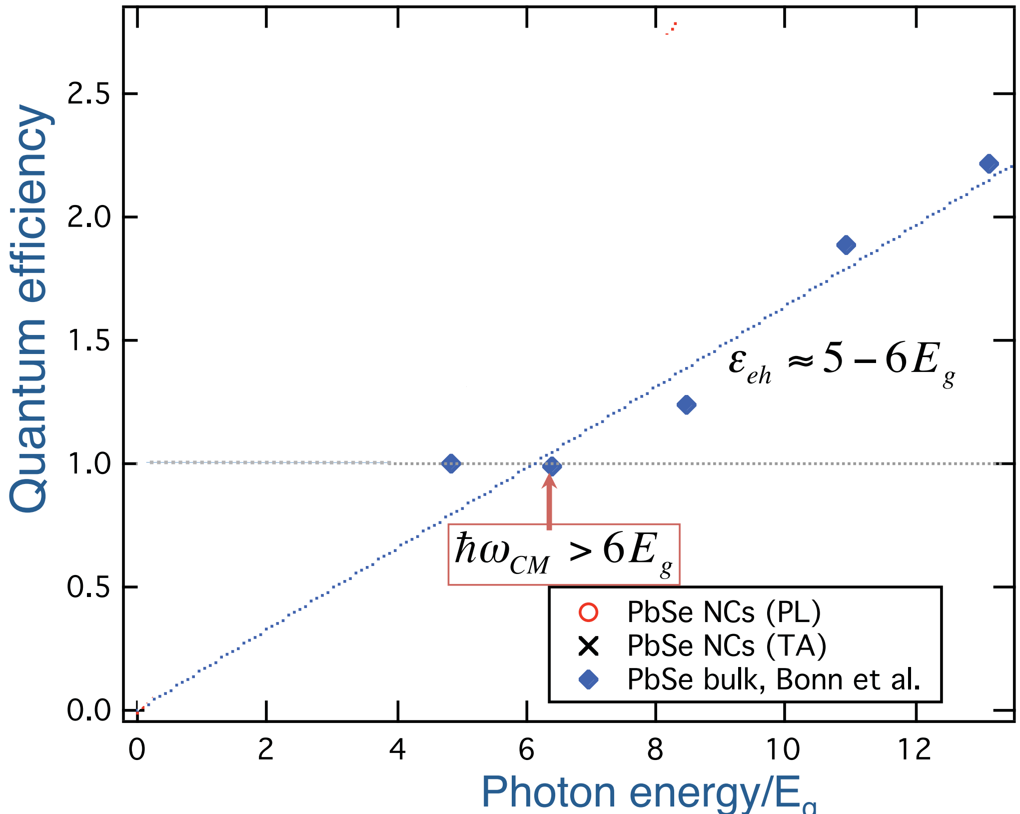
Can reduce $\langle E_{ph} \rangle$

Reduced:

ε_{eh}, $\hbar\omega_{CM}$

R. D. Schaller & V. I. Klimov, *Phys. Rev. Lett.* **92**, 186601 (2004)

Carrier multiplication yields in nanocrystals versus bulk



- ***Impact ionization in bulk PbSe:***

$$QE \approx \frac{\hbar\omega}{\varepsilon_{eh}};\quad \hbar\omega_{CM} \approx \varepsilon_{eh}$$

$$PbSe:\quad \hbar\omega_{CM} \approx \varepsilon_{eh} \approx 6E_g$$

Bulk PbSe (THz): Pijpers, et al. *Nat. Phys.* **5**, 811 (2009)

- ***CM in PbSe NCs:***

$$\hbar\omega_{CM} \approx 2.5E_g$$

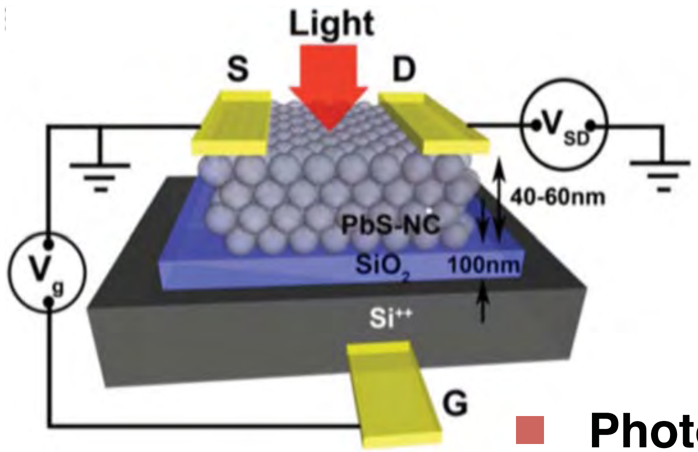
$$\varepsilon_{eh} = 2.3-3.2E_g$$

J. McGuire *et al.*, *Acc. Chem. Res.* **41**, 1819 (2008)
J. McGuire et al. *Nano Lett.* **10**, 2049 (2010)

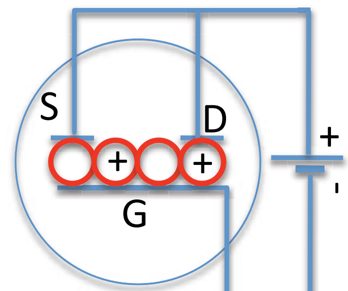
Both CM threshold and e-h pair creation energy are reduced in NCs compared to bulk (if defined in terms of E_g)

Exploratory devices:
Optical field effect transistors (OFETs)

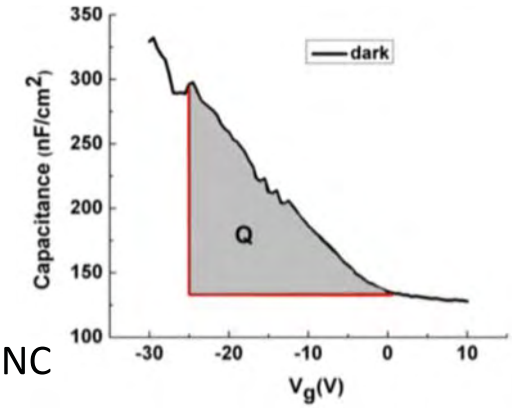
- **PbS NC Optical FET (OFET)**



- **Capacitance measurements (dark)**



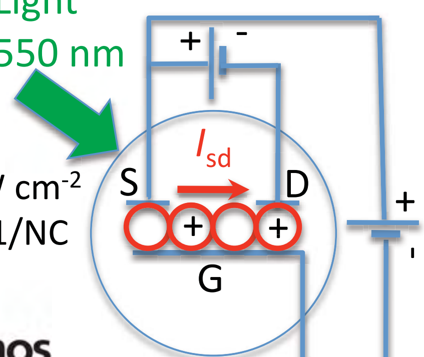
V_g = -25; $n_0 \approx$ 0.1 per NC

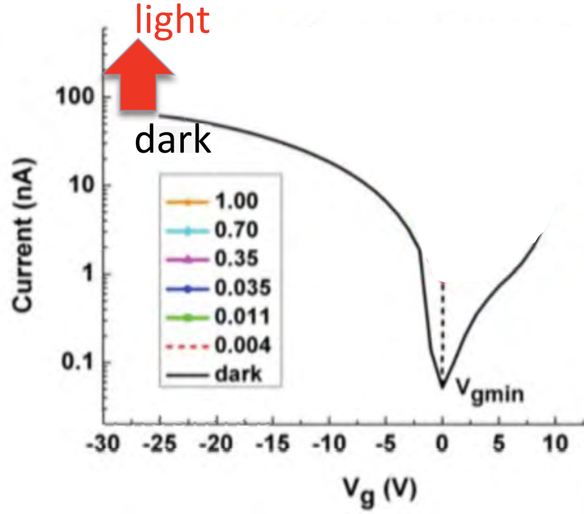


- **Photoconduction vs. dark conductance**

Light
550 nm

W_{max} = 50 μW cm^{-2}
n = p < 0.0001/NC

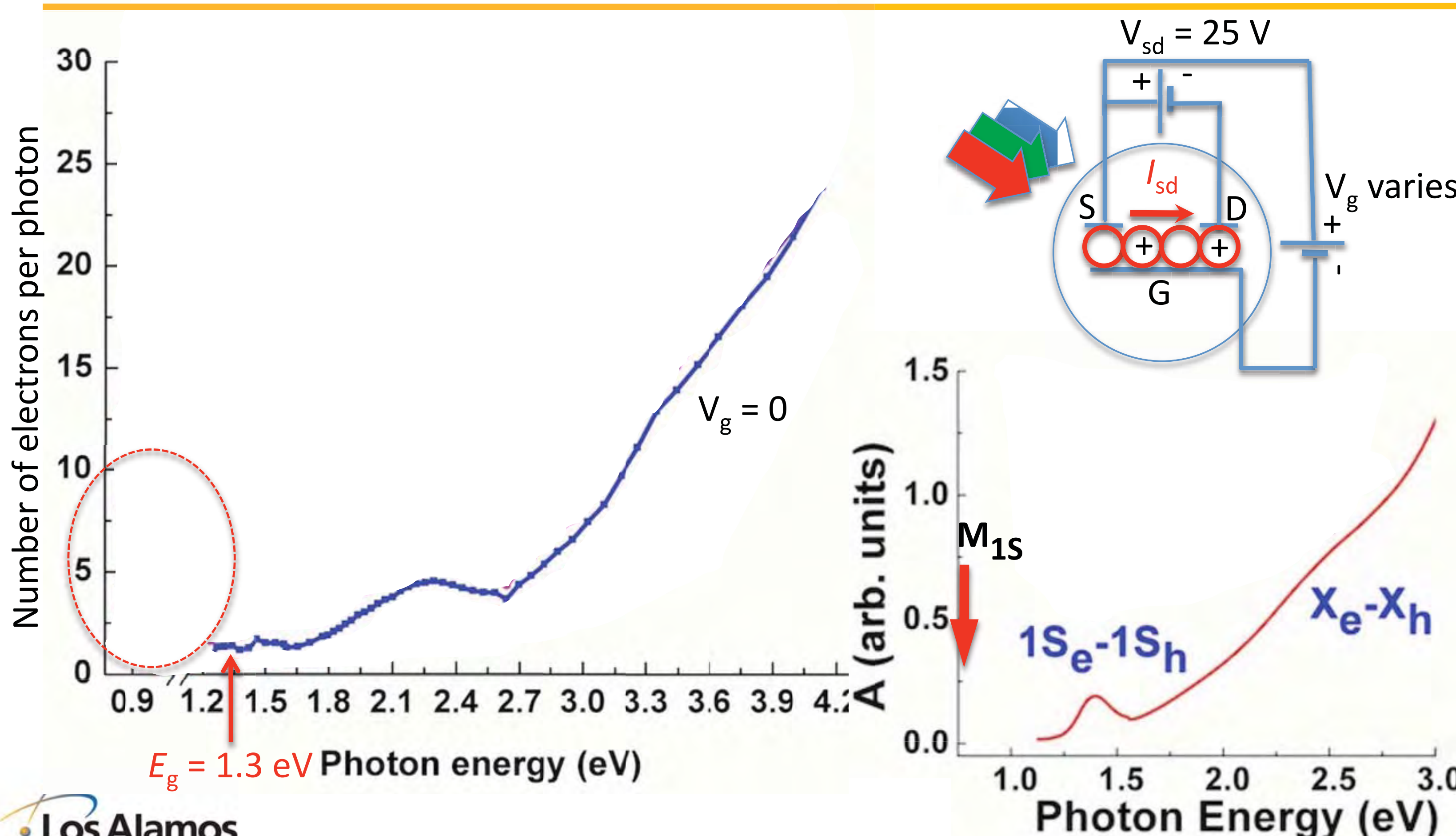




I_{sd} (light) >> I_{sd} (dark)

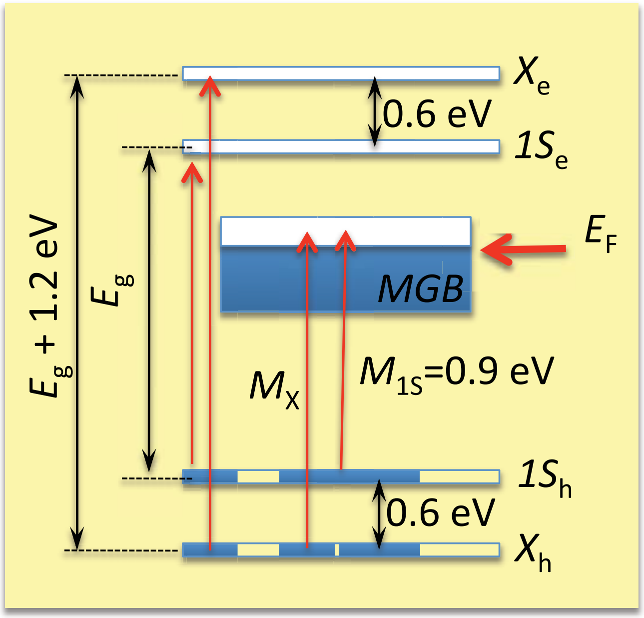
n (light) << n_0(dark)

Mid-gap states are "visualized" in OFET photocurrent

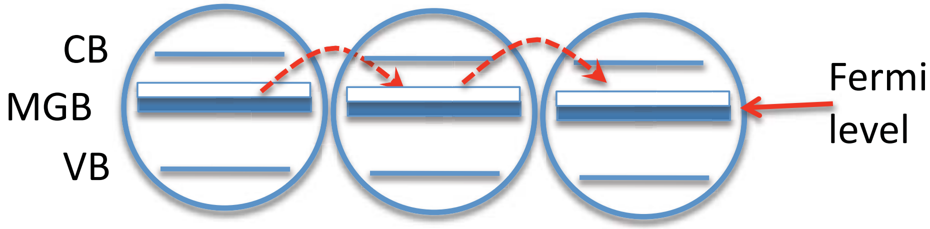


Two mechanisms of charge transport: *“Dark” vs. “light” conductance*

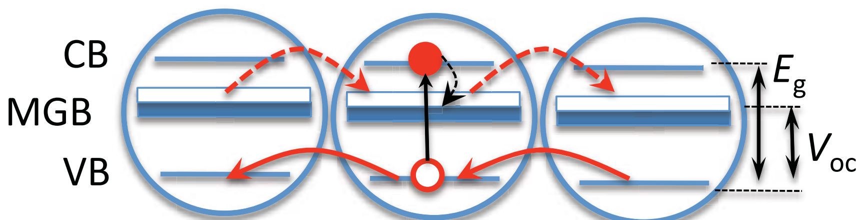
- **Mid-gap band (MGB)**



- **“Dark” conductance: *Mediated by weakly conductive MEG***

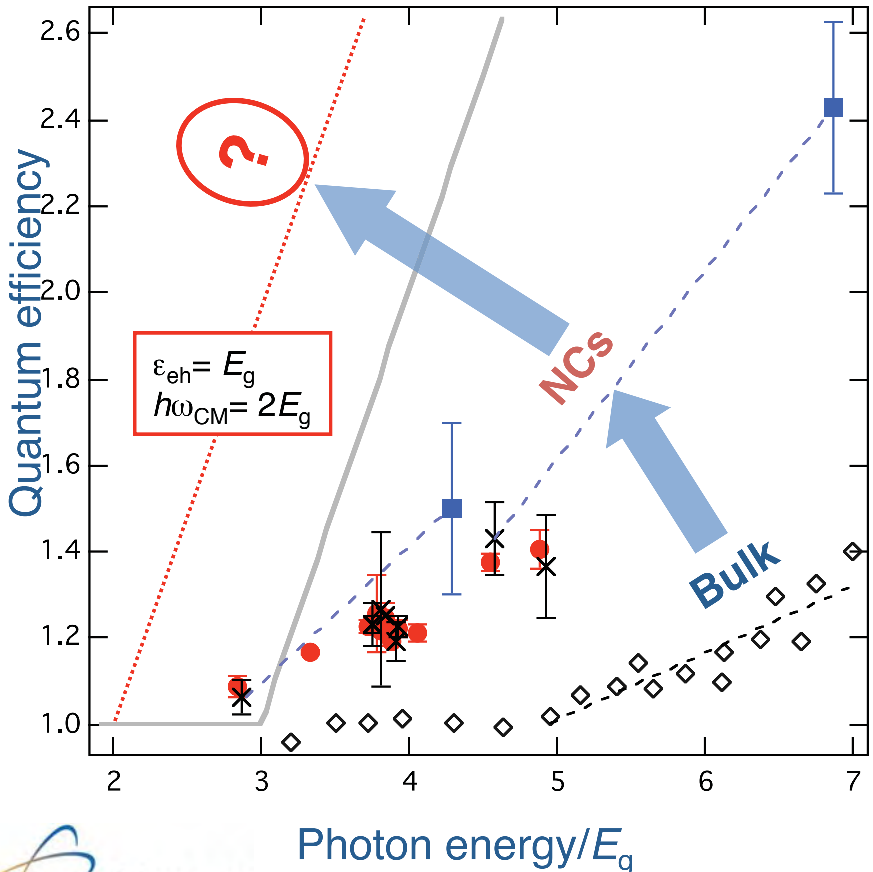


- **Photoconductance: *Dominated by photogenerated holes***



> ➢ *Design principles are different for devices operating in dark (e.g., transistors) and under illumination (e.g., PVs)*
> ➢ *Engineered MGB for, e.g., the realization of ideas of intermediate-band PV cells*

What is next?



- **Understanding CM pathways & competing processes**
  - ***Size/shape/composition effects in CM***
  - ***Role of phonons and surface species***
  - ***New more efficient methods for measuring CM yields***
- **Enhanced properties of NC/ metal nanostructures**
  - ***Practical demonstration of up-conversion via nonlinear ET***
  - ***Demonstration of enhanced excitonic transport***
- **Efficient charge/energy transport in NC assemblies**
  - ***Control of MGB states***
  - ***Reliable demonstration of CM in photocurrent***

CASP results in numbers (FY 2011)

The $3.8M/annum budget has supported:

- 50 working scientists, including over 30 early-career
- $2.23M in matched funds/support
- 30 journal articles (*Science*, *Nature Mater.*, 2 *Phys. Rev. Lett.*, 12 *NanoLett.*, *etc.*)
- 1 provisional patent and 1 patent application
- Over 80 keynote, plenary and invited talks in 13 states and 5 foreign countries
- 2 additional collaborations established (UNM, NMSU)

Colloidal nanostructures – Next frontier in solar

LANL CASP Team
Staff Scientists: V. Klimov, J. Pietryga, I. Robel, H. Htoon, S. Tretiak
Postdocs: J. Stewart, A. Pandey, P. Nagpal, L. Li, B. Diaconescu, C. Galland

Funding: Basic Energy Sciences, Office of Science, US DOE

Materials development in the LANL fuel cell program

R. Borup (MPA-11)

Fuel cells are an important enabling technology for the hydrogen economy and have the potential to revolutionize the way we power the United States and the world, offering cleaner, more-efficient alternatives to the combustion of gasoline and other fossil fuels. Fuel cells have the potential to replace the internal combustion engine in vehicles and provide power in stationary and portable power applications because they are energy-efficient, clean, and fuel-flexible. Although fuel cells have been around since 1839, it took 120 years until NASA demonstrated some of their potential applications in providing power during space flight. Today, the polymer electrolyte membrane (PEM) fuel cell (PEMFC) is the leading fuel cell candidate for automotive applications because it has higher power density, faster start-up and lower cost compared with other fuel cells. While numerous PEM fuel cell automobiles have been demonstrated, fuel cell vehicles still require improvements for widespread commercialization. The cost and durability of PEM fuel cells are the major barriers to the commercialization of these systems for stationary and transportation power applications.

Los Alamos National Laboratory performs applied research and development in fuel cells in support of the DOE Fuel Cell Technologies Program. This research supports the commercialization of PEM fuel cells primarily concentrating on the transportation market. The goal of this work is to execute the enabling science that will produce fuel cell powered vehicles, which will help reduce the United States dependence on foreign oil. A critical component of this research is to help industrial developers further technology towards commercialization, (as this DOE program is an applied program) we will discuss LANL/industrial developer interactions.

The research conducted by Los Alamos addresses the primary barriers of cost and durability for commercialization with component level research conducted in precompetitive areas. A primary challenge in terms of cost for large-scale commercialization, is the electrode use of noble metal (platinum). To address this challenge, we have projects which explore reducing the cost of noble metals by: **Thrifting** – *use less platinum and more other less costly metals,* **Activating** – *increasing the activity of platinum so less can be used,* **Utilize** – *design better electrodes so that the platinum is better utilized,* **Stabilize** – *design better supports to increase the stability (durability) of platinum and enhance the platinum performance,* **Replace** – *develop new electrocatalysts which do not use platinum or other noble metals*. These Los Alamos DOE competitively funded projects will be presented which address the critical cost barrier due to use of noble metal.

Materials Development in the LANL Fuel Cell Program

Presented by: Rod Borup

Fuel Cell Program Manager, MPA-11

Divisions: MPA, MST (LANSCE), C, T

Team consists of electrochemists, chemical engineers, mechanical engineers, theorists, material scientists, and chemists.

UNCLASSIFIED

The state of fuel cells

Fuel Cells for Stationary Power, Auxiliary Power, and Specialty Vehicles

The largest markets for fuel cells today are in stationary power, portable power, auxiliary power units, and forklifts.

~75,000 fuel cells shipped worldwide

~24,000 fuel cells shipped in 2009
(> 40% increase over 2008)

Fuel cells can be a cost-competitive option for critical-load facilities, backup power, and forklifts.

- GM, Toyota, Honda, Nissan, Hyundai and other Global Auto companies are all committed to moving forward on fuel cells, and are well aware of the remaining barriers
- National lab roles include 'fundamental' understanding, materials development, characterization

Fuel Cells for Transportation

In the U.S., there are currently:

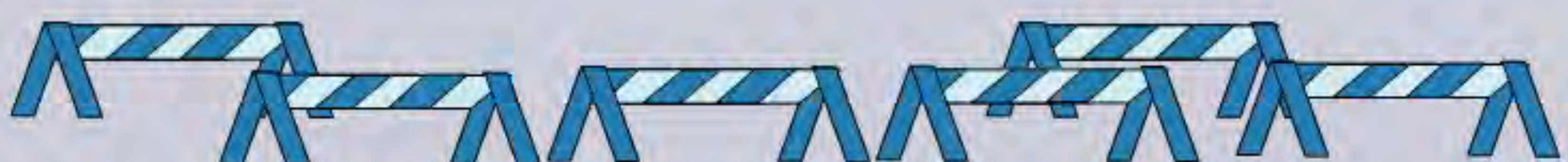
- *> 150 fuel cell vehicles*
- *~ 15 active fuel cell buses*
- *> 50 fueling stations*

Sept. 2009: Auto manufacturers from around the world signed a letter of understanding supporting fuel cell vehicles in anticipation of widespread commercialization, beginning in 2015.

Challenges to Fuel Cell Commercialization

- **Durability**
- **Cost**
- Performance
- Air Management
- System Thermal and Water Management
- Water Transport within the Stack
- Start-up/Shut-down Operation

***Nancy L. Garland, 2008 DOE Hydrogen Program Merit Review and Peer Evaluation Meeting, June 9, 2008**



Los Alamos Hydrogen and Fuel Cell program: DOE EERE funded

Fuel Cells R&D Focus on Cost and Durability

Major Components

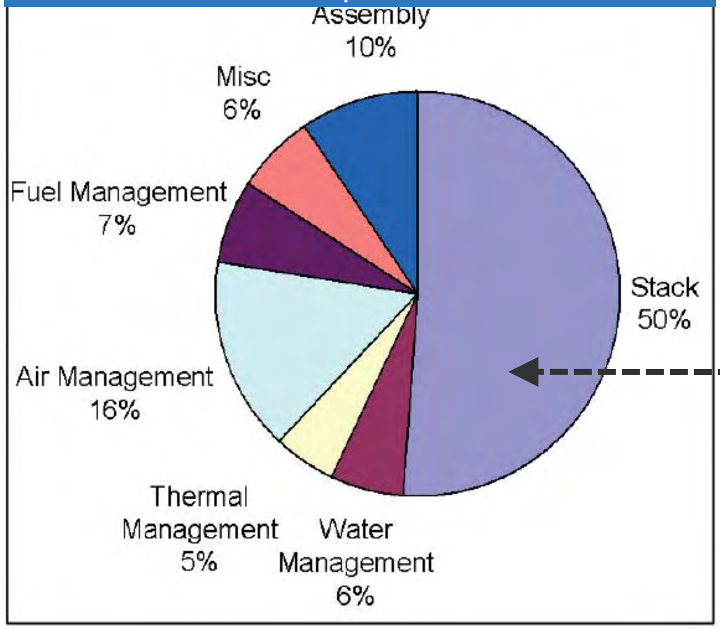
- Fuel cell durability (LANL leads 2 projects, subs on 3 projects)
- Electrocatalysis (LANL leads 2 projects, subs on 3 projects)
- High temperature membranes; alternative membranes (LANL leads 1, sub 1)
- Impurity effects on fuel cell performance (LANL leads 1, sub 1)
- Water transport (LANL leads 1, sub 2)
- Portable power (LANL leads 1)
- 2011 Proposals submitted March: 6 LANL Prime, 10 Sub-contractor
- Supporting: Technical assistance, H_2 codes & standards; fuel quality

~ 90% of LANL Fuel Cell and H_2 Funding is awarded through DOE EERE competitive solicitations

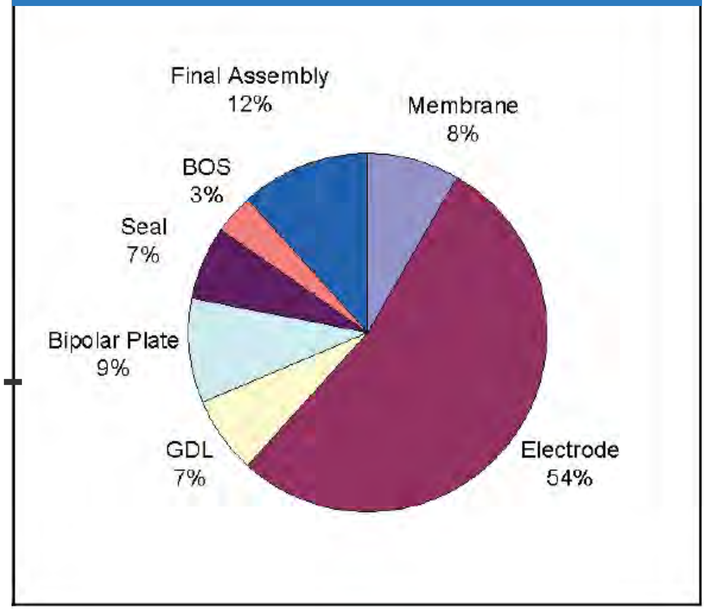
The electrode cost challenge

Approach: Thrift, activate, utilize, stabilize, replace

2008 PEMFC System OEM Cost: $57/kW$_{net\ power}$, $4,560



2008 Cost of Direct-H_2 80 kW Stack: $29/kW, $2,320



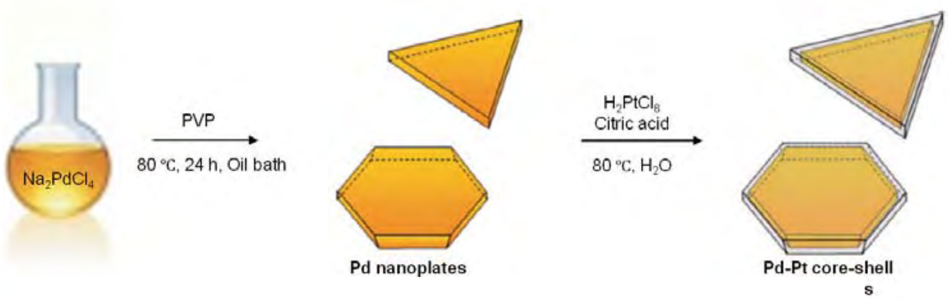
- ***Electrode represents ~ 27% of the total fuel cell system cost!***
  - ***Use of Pt for Oxygen Reduction (ORR) and Hydrogen Oxidation***

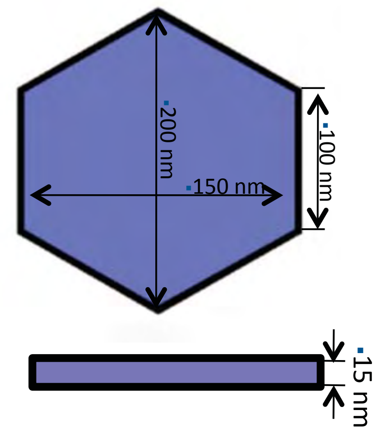
Sinha *et al.*, TIAX LLC, 2009 DOE Hydrogen Program Review Arlington, Virginia, May 21, 2009

New PGM (Pt) catalyst development: Pt on Pd nanoplates

Thrift, Activate

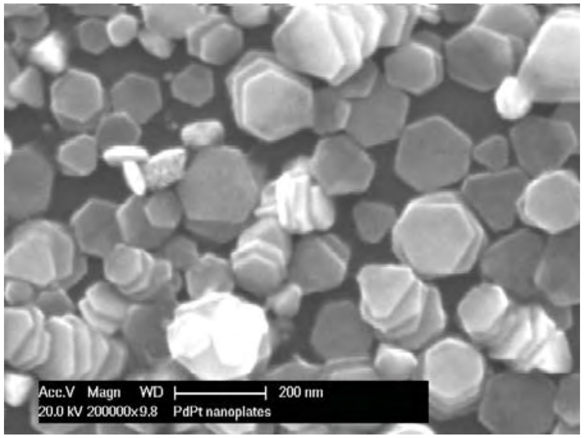
- Pt on Pd has enhanced ORR properties as compared to Pt
  - J. Zhang and R.R.Adzic, **Angew. Chem. Int. Ed.** *2005, 44, 2132-2135*)
- Platelets give more surface area than spheres:





Theoretical surface area:	13 m^2/g M
Pt content for 1 layer:	5.3%
Surface area of Pt:	245 m^2/g Pt

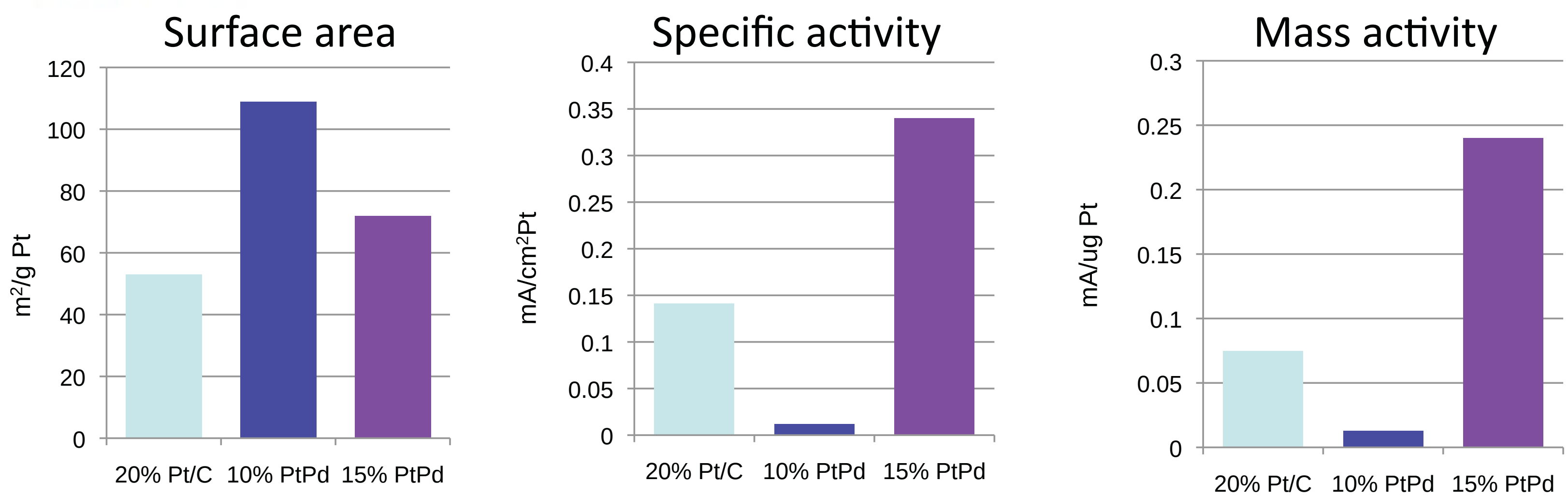
- Samples with different Pt content:
  - 2 layers 10.5%
  - 3 layers 15.5%



SEM image of Pt/Pd Nanoplates

Fernando Garzon et al

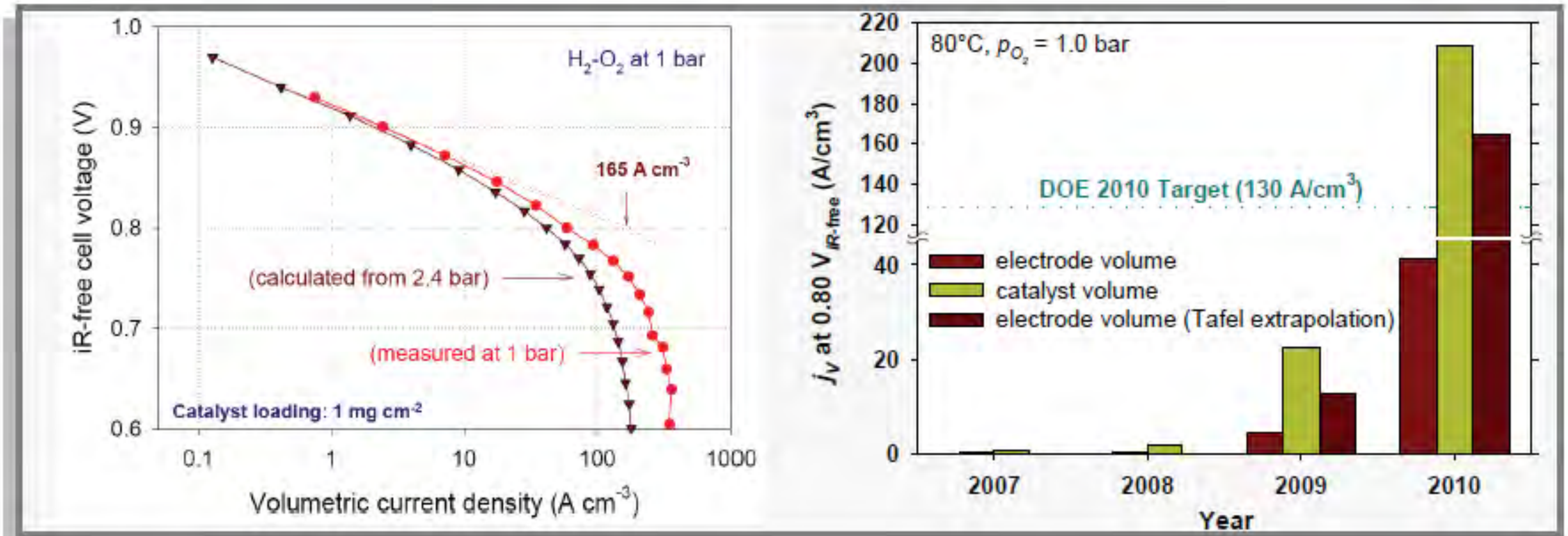
Pt/Pd nanoplates show activity increase over Pt/C



- Surface area for 15% PtPd is higher than Pt/C
- Activity is 2X higher (both specific and mass activity)
- Surface composition has great effect on the activity
    - Surface area increase for 10%PtPd; no activity increase

Non-precious metal catalyst activity increased

Replace



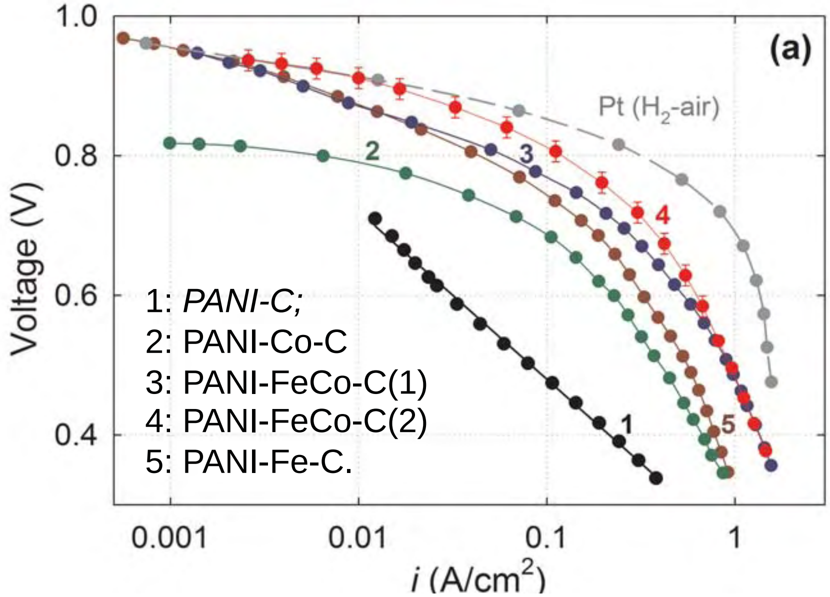
- **High ORR activity reached with several non-PGM catalysts by LANL, including cyanamide-Fe-C catalyst (shown)**
- **Fuel cell performance improved by more than 100 since 2008**
- **Catalyst activity exceeds DOE 2010 activity target of 130 A/cm^3 at 0.80 V**

Piotr Zelenay et al

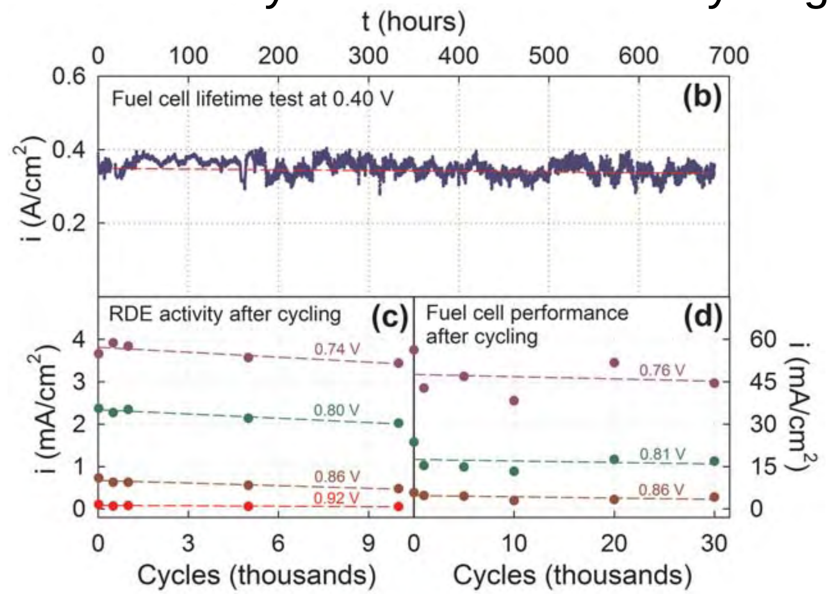
PANI-derived non-PGM electrocatalysts

High performance plus durability

H_2-O_2 fuel cell polarization plots



Durability in Fuel Cells and Cycling

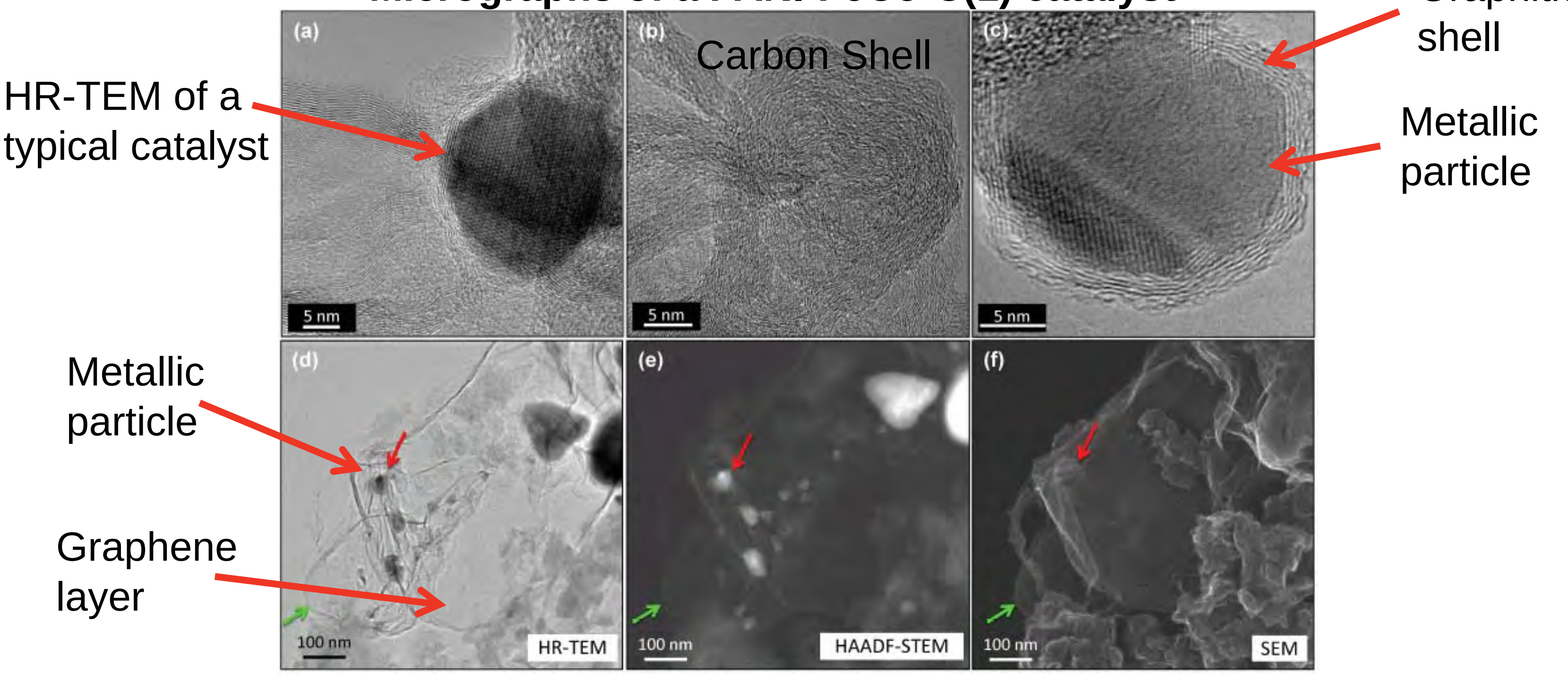


- Polyaniline as a precursor to a carbon-nitrogen template for high-temperature synthesis of catalysts incorporating iron and cobalt.
- Worlds best performing durable non-PGM electrocatalysts

Gang Wu*, et al., Science* ***332, 443 (2011)***

How to improve non-PGM electrocatalysts: Define structure and active ORR site

Micrographs of a PANI-FeCo-C(1) catalyst

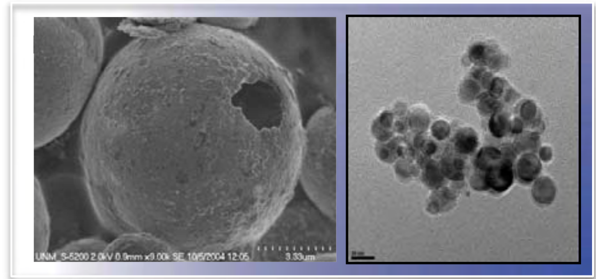


- Identity of active site remains unclear
- Competing models exist
- LANL LDRD project to identify non-PGM catalyst active site

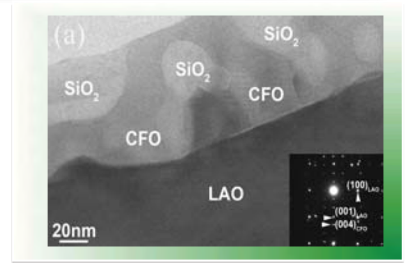
Ceramic supports for Pt (*Activate , Stabilize*)

Replace carbon with conductive ceramic materials

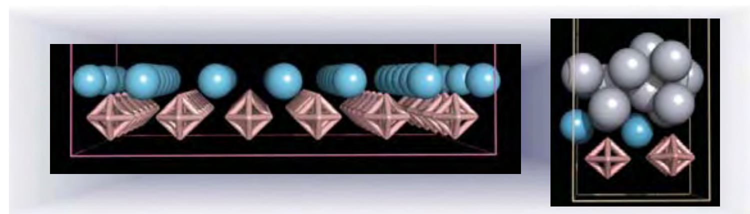
- **Microwave aerosol-through-plasma (ATP) torch synthesis of hexaborides and sub-stoichiometric titanias**
  - Plasma: high temperature/short contact times
  - T > 3500K, t < 0.1 sec



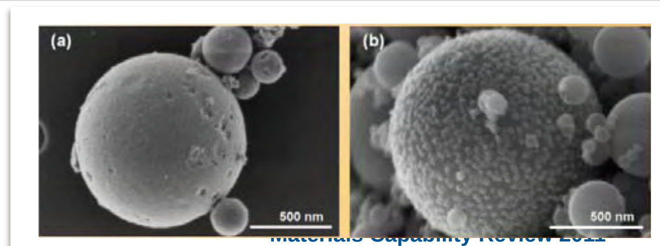
- **Polymer assisted deposition (PAD) for hexaborides and nitrides.**
  - Ceramic materials with high surface areas
  - Films (CVs), powders (bulk catalysts, MEA prep)



- **Modeling to aid experimental effort to provide stability data**
  - Surface/cluster models to predict effects of particle size reduction, conductivity.
  - Study nature of Pt binding sites, interaction energy, etc.



- **Conductive NbO_2 and RuO_2 supports**
  - Spray pyrolysis methods to prepared conductive metal oxide supports



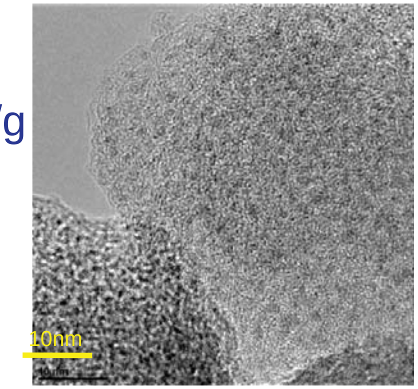
Nano-sized molybdenum nitride supports

Promising surface areas and conductivity

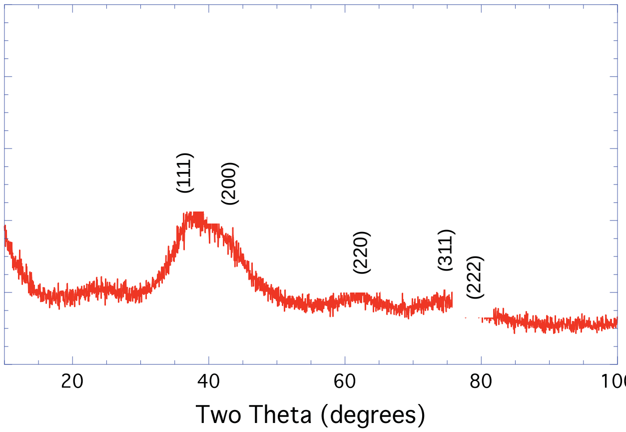
Mo$_2$N synthesis

- Polymer Assisted Deposition:
  - 1 – 2 nm sized crystallites
  - BET surface areas ~ 300m^2/g typical, up to 500m^2/g
  - Electrical conductivity: Mo$_2$N roughly ½ of XC-72R
  - Pt/Mo$_2$N catalysts exhibit activity comparable to Pt/C

TEM

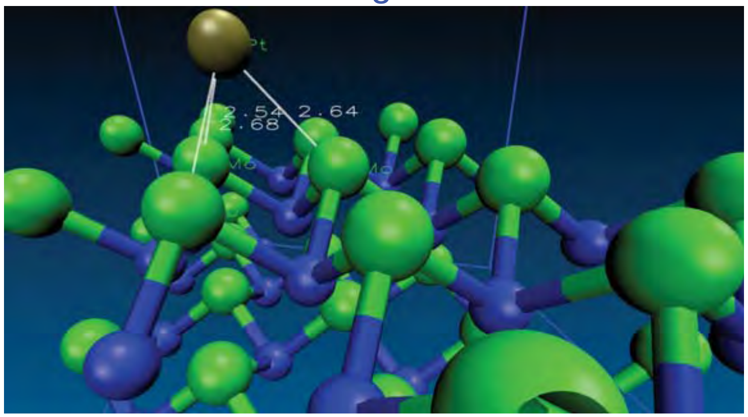


Pt XRD – Particle Size Analysis

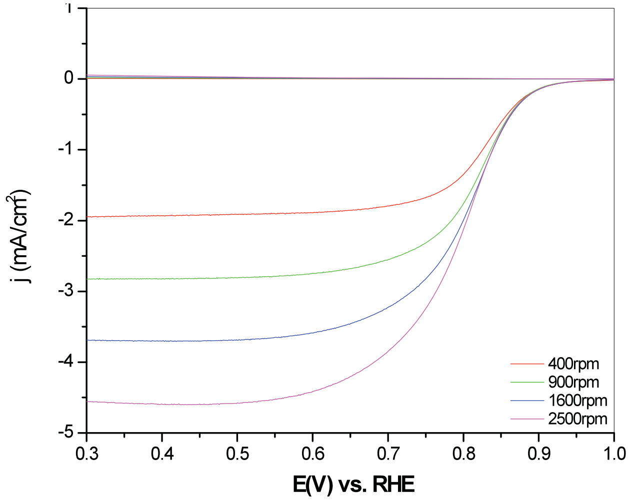


- Modeling nucleation and growth of Pt indicate strong Pt interaction with Mo-N surfaces

Favored Pt bonding sites modeled



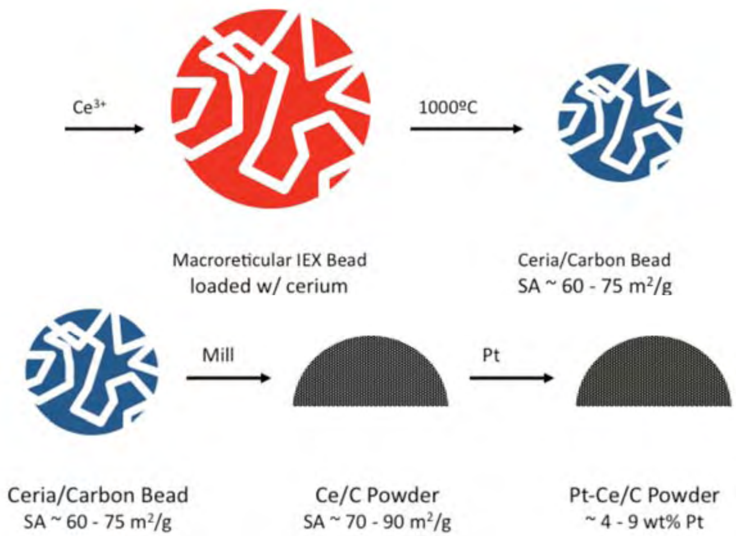
RRDE: 20 wt% Pt dispersed onto Mo$_2$N



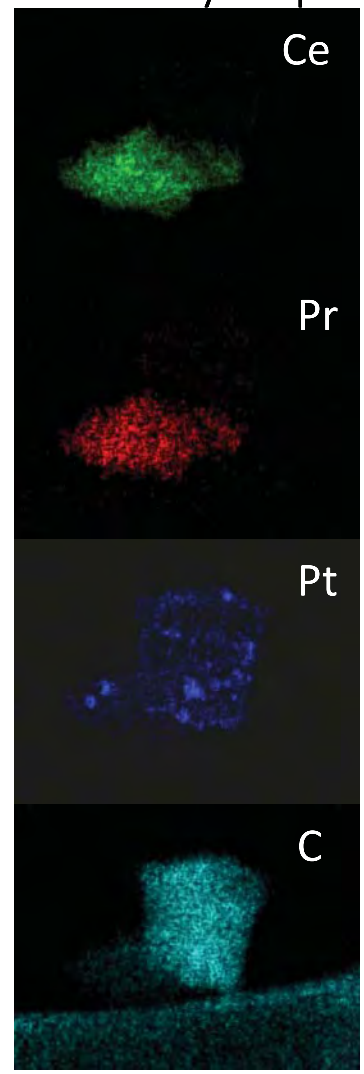
- Similar activity to Pt/C

Pt/MOx/C catalysts: Provide improved catalysis and free radical scavenging

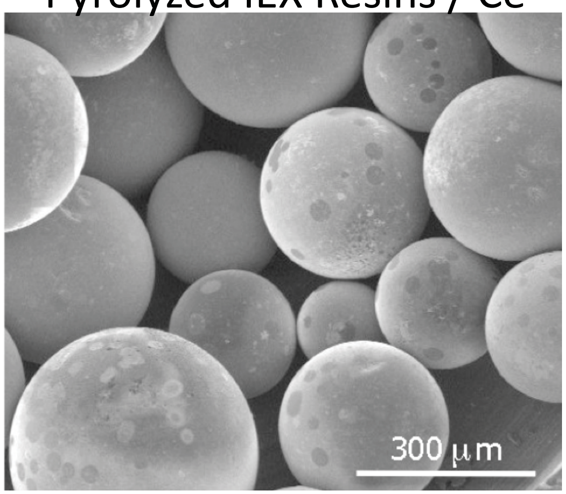
- **Pt supported on metal oxides**
- **Absorb precursor solutions onto carbonaceous sorption media then pyrolyze to form nanocomposites**
- **Ceria, Gd doped Ceria, and Pr doped Ceria supports synthesized**
  - Pt deposition from acetone H_2PtCl_6



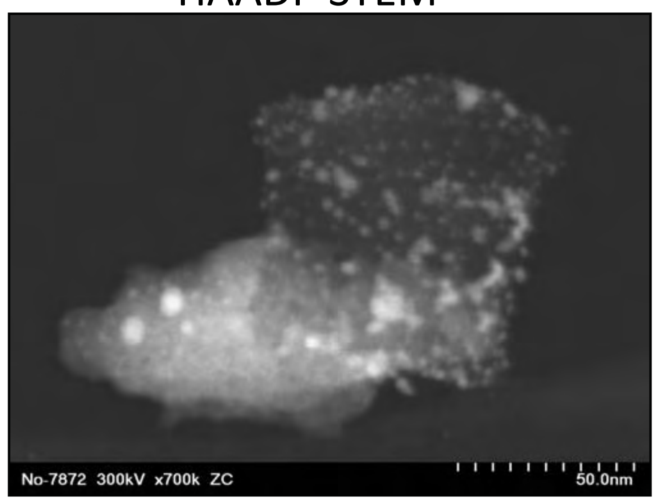
EDS-X ray Maps



Pyrolyzed IEX Resins / Ce



HAADF-STEM

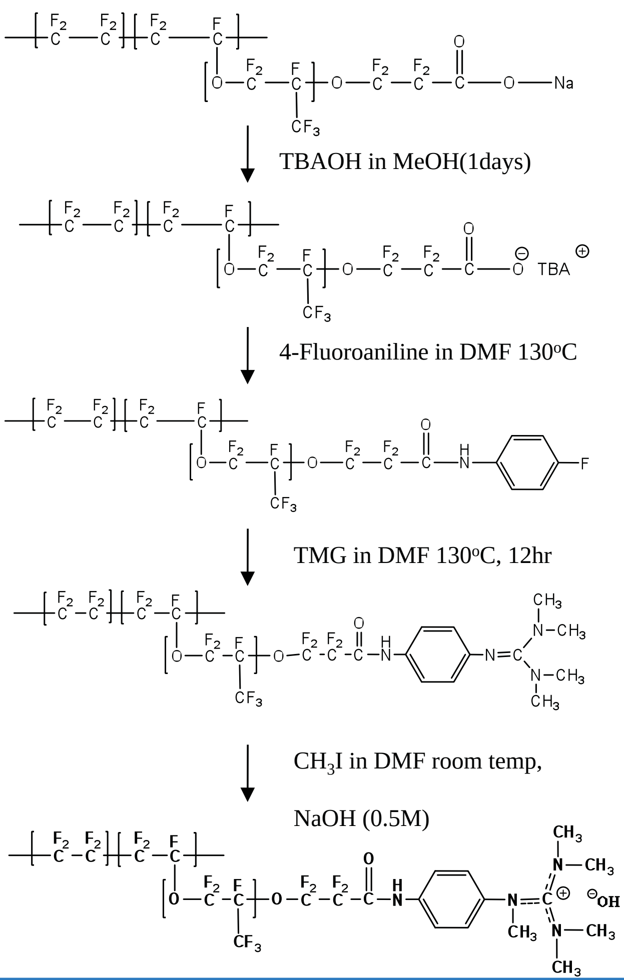


Alkaline electrolytes: Guanidinium based perfluorinated ionomers

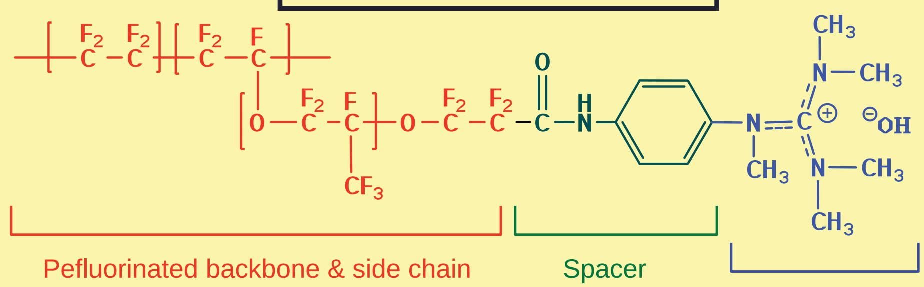
Replace

Non-noble metals more stable, active in alkaline media Y.S. Kim et al

Synthesis



*Perfluorinated ionomer**



IEC= 0.74 meq./g, σ = 20 mS/cm (80ºC)

Perfluorinated backbone & side chain

- **Provide high gas permeability**
- Inert to electro-catalytic reaction
- Chemically and oxidative stability

Spacer

- **Cation stability by increasing electron density**

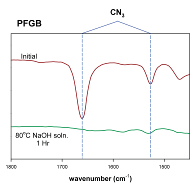
Cation

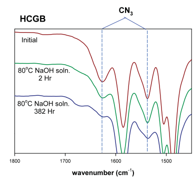
- Cation stability by resonance structure
- **Provide highly active catalyst-electrolyte interface**

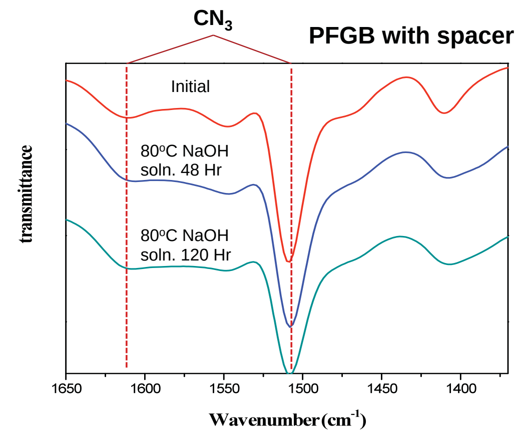
** LANL patent pending (2011)*

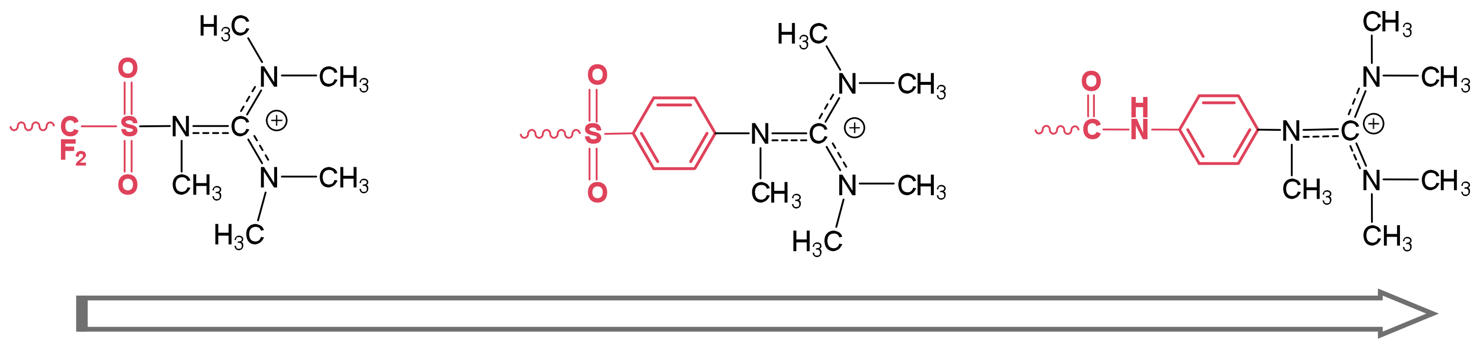
Electron donating spacer stabilizes alkaline ionomer

FTIR of New and Aged Alkaline Ionomers









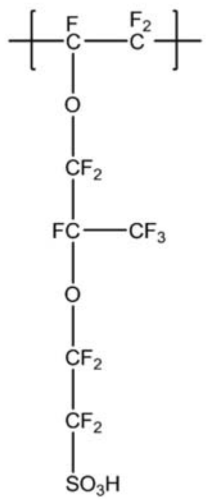
Local electron density & stability

- **Cation stability of perfluorinated AEM significantly improved by electron donating spacer**
- This is a proof of principle for utilizing perfluorinated ionomer for AMFC
- Fuel cell testing underway. Membrane conductivity currently ~ ½ of proton conductors – need improved durability for thinner membranes, lower conductivity Ohmic losses

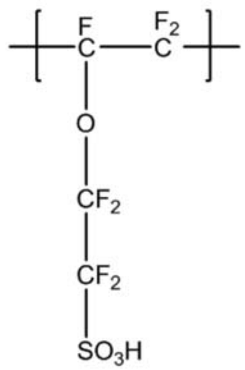
Ionomer and electrode structure effects on catalyst utilization and durability

Perfluorinated sulfonic acid polymers

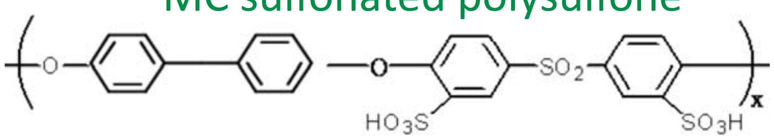
Long side chain (LSC) PFSA (Nafion®)



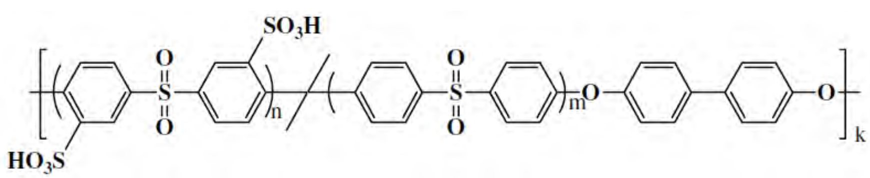
Short side chain (SSC) PFSA (Aquivion)



MC sulfonated polysulfone*



Sulfonated poly(arylene ether sulfone)s (*BPSH*)



Understand mechanisms behind loss of performance

MEA Variables:

- Catalysts
  - Loadings
  - Supports (graphitization/surface area)
- Membranes
  - Nafion / Reinforced / Stabilized
  - Hydrocarbon (ionomer analysis)
- Electrode Layer Ionomers
  - Long side chain (LSC)
  - Short side chain (SSC)
  - Stabilized/ Un-stabilized Nafion
  - Nafion – digested after degradation
- Electrode Structure
  - Solvent effect
  - (water/iso-propanol, glycerol)

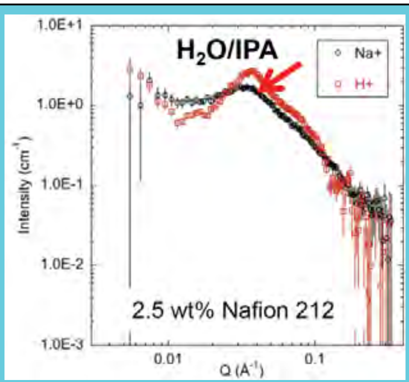
Utilize
Stabilize

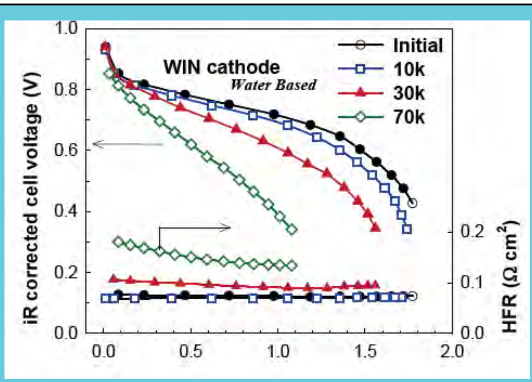
R Borup, et al

Electrode structure shows effect on catalyst durability

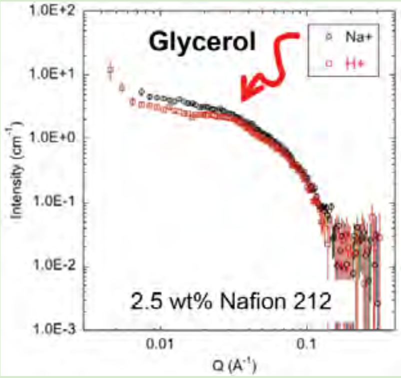
Utilize
Stabilize

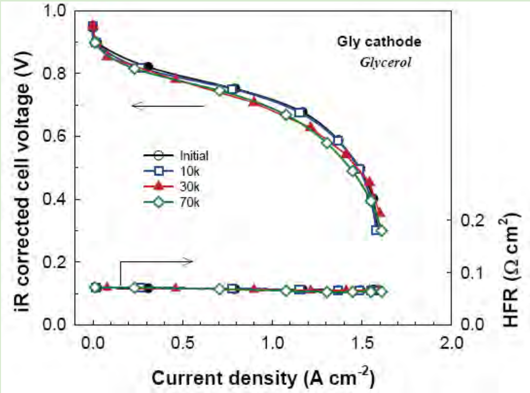
Determine effect of electrode processing on durability



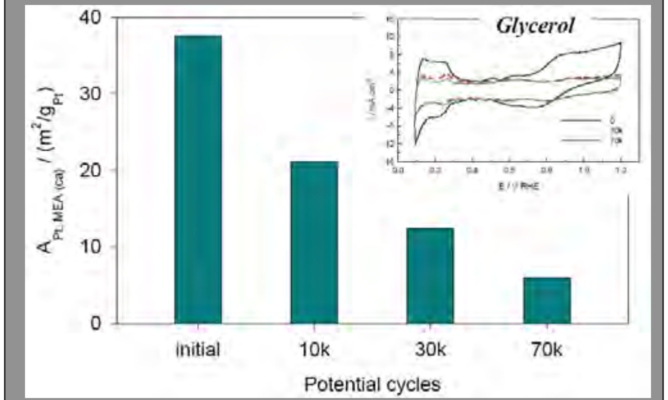


Ionomer ordering in water/alcohol → **Poor durability**

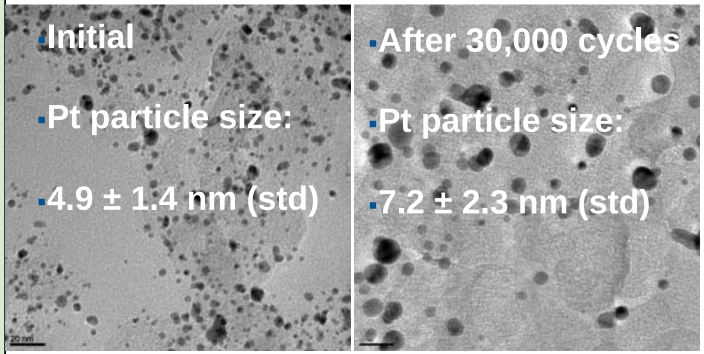




Lack of ordering in glycerol → **High durability – exceeds 30,000 cycle target**

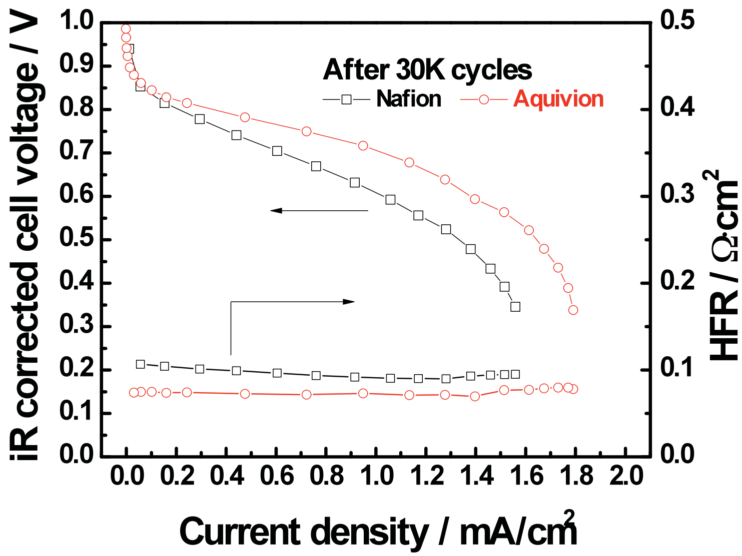


Catalyst degradation is complex – major loss in ECSA with negligible loss in performance

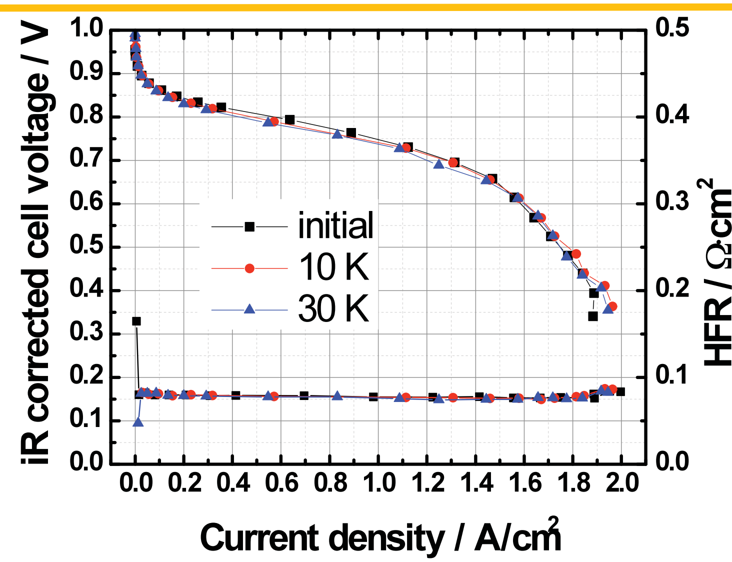


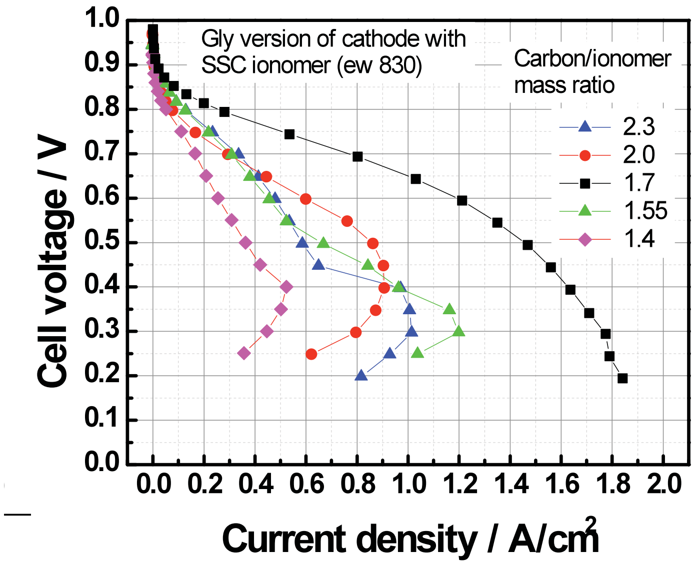
C. Johnston, et al

Short-side-chained (SSC) ionomers show improved durability



- SSC Ionomer shows improved durability for catalyst cycling durability AST
- Reduction in ionomer content yields best performance
  - Demonstrates optimized carbon/ionomer ratio depends on electrode structure
- Disconnect between ECSA and performance for Pt/C (similar to solvent effect)

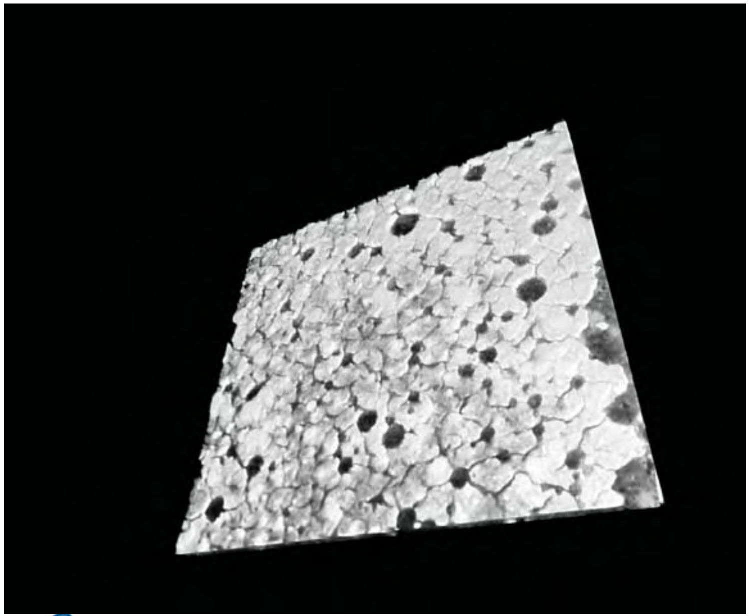




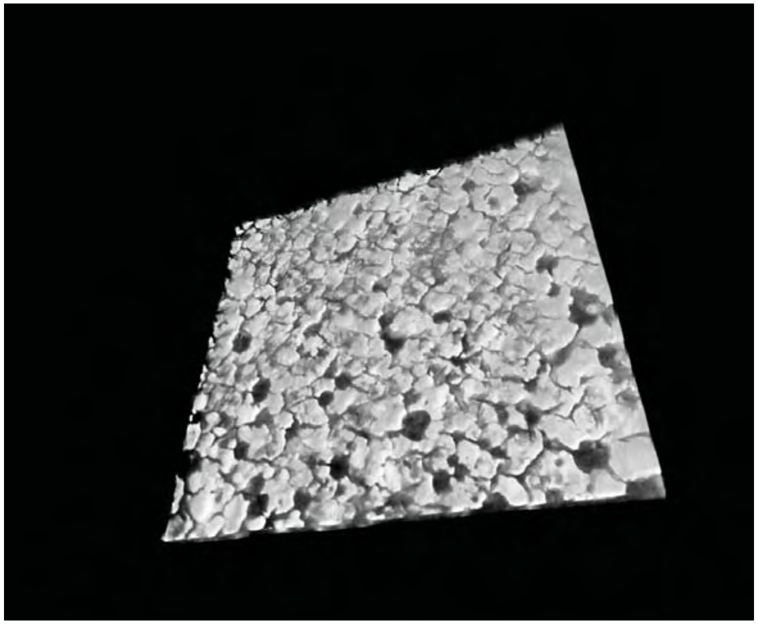
Industrial support: Advanced materials/ component characterization

- LANL supports industrial developers with capabilities not typically available in industry (especially at small developers)
- Example: X-ray tomography of new/used Membrane/Electrode/Assemblies
  - Use to optimize initial structure, quality control, durability aspects

MEA New



After Corrosion Protocol 1.2V



Carbon corrosion at crack edges

2011 solicitation partners and other current technical partners

2011 Solicitation

- General Motors
- Nissan Motor Company
- Hyundai Motors Company
- 3M
- Delphi Automotive
- W.L. Gore
- BASF
- SGL Carbon, GMBH
- Automotive Fuel Cell Cooperation (AFCC)
- Ballard Power Systems
- Ceramatec Inc.
- IRD Fuel Cells
- Tetramer
- KWJ Engineering
- Donaldson
- Akron Polymer Systems
- Energy Conversion Devices
- ClearEdge Power
- PBI Performance Products
- Treadstone Technologies Inc.
- Engineered Fibers Technology
- Lawrence Berkeley Nat. Lab
- Oak Ridge Nat. Lab
- Nat. Inst. of Standards and Tech.
- Case Western Reserve Univ.
- Carnegie Mellon University
- Univ. of Texas (Austin)
- Univ. of New Mexico
- University of Rochester
- University of Waterloo
- Simon Fraser University
- Cornell University
- Northeastern University
- University of Tennessee
- National Research Council of Canada
- Univ. South Carolina
- University of California (Irvine)

2008 Solicitation

- Nuvera Fuel Cells
- UTC
- Ion Power
- Argonne Nat. Lab
- Brookhaven Nat. Lab
- Sandia Nat. Lab
- NREL (National Renewable Energy Lab)
- Virginia Tech.
- Univ. of Illinois
- Univ. of Cal (Riverside)
- **(Incomplete List)**

Summary

Fuel Cell program addresses cost and durability

- **Electrode cost is a primary challenge for both**
  - Thrift and Activate
    - New Pt structures
    - Pt augmentation
  - Stabilize
    - Durable supports
    - New support interactions
  - Replace
    - Non-PGM catalysts based on cyanamide-Iron-carbon
    - Derived from polyaniline with iron and cobalt
    - New electrolytes for alkaline
  - Utilization
    - Electrode structure
    - Processing effects
- **Durability**
    - Understand fundamental degradation mechanisms
- **Support industrial developers**
  - Provide characterization capabilities not typically available in industry

Thanks to

- **U.S. DOE -EERE Fuel Cell Technologies Program for financial support of this work**
  - Technology Development Manager: Nancy Garland

- **The LANL Fuel Cell Team and all of its partners**

Colloidal nanomaterials for light harvesting applications

A. Pandey, L. Li, B. P. Khanal, H. Tsai, H. Wang, J.M. Pietryga, V.I. Klimov (C-PCS)

We describe our efforts towards the development of novel, inexpensive solution-processable nanomaterials to enable cost-effective capture of solar radiation. In this poster, we address the synthesis of novel semiconductor nanocrystals (NCs) and metal-semiconductor hybrids. Copper indium sulfide (CIS) NCs have a band-gap energy well suited for applications in thin-film photovoltaics. We have developed a new synthesis of CIS NCs using a scalable, virtually waste-free approach. The growth of an inorganic shell boosts emission quantum yields to 90%, indicating excellent surface passivation. We study the role of defects in these NCs and demonstrate that the emission occurs through the recombination of a conduction-band electron with a localized hole. These results help to rationalize a significant difference between electron and hole conductivities in CIS nanocrystal films. In a separate effort, we synthesize and study hybrid structures that combine semiconductor NCs with nanoscale metals. Metal plasmons can significantly modify the behavior of proximal NCs, e.g., enhancing absorption and modifying emission and energy transfer rates. The goal of our study is to develop solution-processed hybrid materials with optical responses useful for light harvesting. We will discuss the synthesis and optical properties of tunable hybrid structures with a focus on novel features associated with strong semiconductor-metal coupling.

A comparative study of carrier multiplication yields in PbSe and PbS nanocrystals

J. Stewart, C-PCS; A. Midgett, NREL, CU; L. Padilha, C-PCS; D. Smith; NREL; J. Pietryga, J. Luther, C-PCS; M. Beard, NREL; A. Nozik, NREL, CU; and V. Klimov, C-PCS

We present recent results in which we compare Auger-recombination lifetimes and carrier multiplication (CM) yields for strongly confined PbSe and PbS nanocrystal quantum dots (QDs). QDs made of PbX are attractive candidates for third-generation solar cells because of their narrow band gaps, large Bohr exciton radii, and good natural abundance. In this work, we study a large collection of samples using two different experimental techniques (photoluminescence up-conversion and transient absorption) spread over two different institutions. We observe that the biexciton Auger lifetimes are similar for PbSe and PbS QDs with matching band gap energies (Eg), indicating similar strengths of Coulomb interactions. Given other parallels between the materials, one would predict similar CM efficiencies; however, we find that these two types of the QDs exhibit strikingly different CM trends. Specifically, we observe that for all PbSe QDs studied, the quantum yield (QY) of photon-to-exciton conversion increases with increasing the ratio of the photon energy ($h\nu$) to Eg. PbS QDs, however, follow an initial increase and then the QY drops above a certain values of $h\nu$/Eg. We analyze these results in the context of known trends for phonon emission rates in bulk PbSe and PbS as well as CM rates expected based on the measured Auger recombination lifetimes.

Assessment of silicon nanowire architecture for PV application

S. Dayeh, J. Yoo, D. Perea, S. Picraux (MPA-CINT); I. Campbell (MPA-11)

1D semiconductor nanowire arrays have great potential for improved photovoltaic performance and cost reduction over their bulk counterparts with sufficiently thin absorption lengths. This is due to:
(i) improved absorption from diffuse light scattering in nanowire arrays, (ii) short collection lengths of minority carriers that are radially separated and collected normal to the light absorption direction, (iii) flexibility of integration on a variety of carrier substrates.

The goals of this project are to experimentally determine whether silicon nanowire solar cells can achieve performance close to that of crystalline silicon cells but with the much lower cost of thin film, amorphous Si solar cells. Here, we provide intuitive device physics models for radial nanowire solar cells and demonstrate the fabrication of radial pillar arrays over large areas on thin Si substrates using a CMOS fab-compatible process. We also provide the first direct measurement of critical thicknesses (axial and radial strain relaxation) in Ge/Si core/shell heterostructures of potential in fabricating Tandem cells.

Controlling electrode morphology to improve fuel cell performance and durability

C. Welch, E.B. Orler, MST-7; R. Hjelm, LANSCE-LC; C. Johnston, Y.S. Kim, B. Choi, MPA-11; A. Labouriau, MST-7; N. Mack, C-PCS; M. Hawley, MST-8; K. More, ORNL

Polymer electrolyte fuel cells show promise as a clean and competitive energy source for automotive applications, but a major barrier to their commercialization is the low performance durability of the Pt-based electrodes to potential cycling. While recent efforts to address this issue have focused on preventing Pt particle growth, the impact of changing the catalyst/polymer electrode structure has been largely unexplored. Our research shows that polymer-solvent interactions at play during electrode fabrication can strongly influence fuel cell performance and durability. Using small-angle neutron scattering (SANS) and NMR, we have investigated the morphology of polymer dispersions used to create electrodes. The results are interpreted in light of polymer-solvent thermodynamics.

Nanoscale ceramic catalyst support synthesis

E. Brosha, L. Elbaz (MPA-11); K. Blackmore, A. Burrell (MPA-MC); N. Henson (T-1)

Catalyst support durability is currently a technical barrier for commercialization of polymer electrolyte membrane (PEM) fuel cells, especially for transportation applications. Degradation and corrosion of the conventional carbon supports leads to losses in active catalyst surface area and, consequently, reduced performance. The benefits of the use of carbon-supported catalysts to drastically reduce Pt loadings from the early, conventional Pt-black technology are well known. Of direct relevance to this present work, are the investigations into Pt particle growth in PEM fuel cells, and subsequent follow-on work showing evidence of Pt particles suspended free of the support within the catalyst layer, Further, durability work has demonstrated the detrimental effects of potential cycling on carbon corrosion and the link between electrochemical surface area and particle growth. To avoid the issues with carbon degradation altogether, it has been proposed by numerous fuel cell research groups to replace carbon supports with conductive materials that are ceramic in nature. Intrinsically, these many conductive oxides, carbides, and nitrides possess the prerequisite electronic conductivity required, and offer corrosion resistance in PEMFC environments; however, most reports indicate that obtaining sufficient surface area remains a significant barrier to obtaining desirable fuel cell performance. Ceramic materials that exhibit high electrical conductivity and necessary stability under fuel cell conditions must also exhibit high surface area as a necessary adjunct to obtaining high Pt dispersions and Pt utilization targets. As a result, the major aim of this work is to develop support materials that interact strongly with Pt, yet sustain bulk-like catalytic activities with very highly dispersed particles. This latter aspect is key to attaining the 2015 DOE technical targets for platinum group metal (PGM) loadings (0.20 mg/cm^2).

Membrane materials for energy applications

K.A. Berchtold, R.P. Singh, K.W. Dudeck, V.A. Kusuma (MPA-MC); D. Yang, C.F. Welch (MST-7)

There is compelling need to develop novel separation methods to improve the energy efficiency of synthesis (syn) gas processing operations including H_2 and H_2/CO production to meet power, chemicals, and fuel producer needs, as well as carbon capture and removal of other undesirable syn gas impurities. To be technically and economically viable, a successful separation method must be applicable to industrially relevant gas streams at realistic process conditions and compatible with large gas volumes. Our team is developing H_2 selective polymer-based membranes for syn gas separations at elevated temperatures.

The overarching goal of this work is to develop and demonstrate polymer-based membrane chemistries and structures that achieve the critical combination of high H_2 permselectivity, and chemical and mechanical stability. Polybenzimidazole (PBI)-based polymer chemistries have been identified as exceptional candidates for H_2 purification and CO_2 capture from syn gas. Our work involving this class of selective barrier materials is focused on correlating the effects of PBI molecular structure and chemistry variations on permselectivity and material properties at elevated temperatures, and ultimately developing methods to effectively translate those developed materials into commercially viable membrane platforms and separation schemes. Free volume or cavity size and its distribution ensuing from molecular and macromolecular characteristics (such as: chain packing, intra- and inter-molecular interactions, and orientation) govern the permselectivity character of these materials. To understand and ultimately manipulate the free volume architecture of these materials for gas transport, a more comprehensive understanding of these controlling factors and their qualitative and quantitative influence on material free volume is under development. We will discuss our work aimed at inducing free volume changes via tailored monomer/polymer synthesis and membrane fabrication protocols and the cross-correlation of those manipulations with free volume and permselectivity information obtained via positron annihilation spectroscopy and gas permeation experiments.

Chemical H_2 storage research at LANL

A. Burrell, B. Davis, H. Diyabalanage, T. Nakagawa, T., Semelsberger; R., Shrestha (MPA-MC); J. Gordon, F. Stephens; A. Sutton (C-IIAC); K. Ott (SPO-AE)

Los Alamos co-leads the Chemical Hydrogen Storage Center of Excellence for the Department of Energy Office of Energy Efficiency and Renewable Energy. Substantial progress was made developing chemical hydrogen storage materials through collaboration between industry, government, and national labs. By administering a well-crafted down-select process, the large pool of candidate materials was quickly reduced to focus our effort. In the last year, focus has shifted to fluid phase materials, particularly of ammonia borane (AB), because of its high gravimetric capacity of H_2 (~19 wt. %), lightweight, and developed thermolytic release chemistry. Progress characterizing the ionic liquids and resulting AB compositions will be detailed. In addition, last year marked the publication of a new single pot regeneration of spent AB fuel with hydrazine. This chemistry will be described and future challenges identified.

In 2009, EERE funded the Hydrogen Storage Engineering Center of Excellence to transition discoveries from the three major CoE (adsorbents, metal hydrides, and chemical) to an operable H_2 delivering system. LANL plays a substantial role in this effort with a focus on chemical hydrogen storage materials. In the last year, a small-scale fluid-phase validation test bed was constructed and used to release H_2 from 5% AB in tetraglyme solution, a surrogate for fluid phase H_2 storage materials in development. This test bed allows for fluid phase materials to be evaluated in a controlled fashion (flow rate, temperature, catalyst bed), with the H_2 gas stream outflow being analyzed in situ (FTIR and microGC) for impurities. Additionally, impurity-adsorbing media can be installed in-line to test efficacy. Using a similar system, the impurities NH_3, borazine, and diborane were quantified from the thermolytic decomposition of solid AB. Also in the last year, a new acoustic fuel gauge sensor has been developed for metal hydride containing tanks (patent filed); the general paradigm may be extended to chemical hydrogen and cryogenic adsorbent tanks as well.

Comparative studies of vortex physics in oxide, iron-arsenide and MgB_2 superconductors

L. Civale, B. Maiorov, N. Haberkorn, M. Miura, F.J. Baca, P. Dowden, T.G. Holesinger, (MPA-STC); J. Kim, M. Jaime, (MPA-CMMS); I. Usov (MST-7); L. Boulaevskii (T-4); T. Katase, H. Hiramatsu, H. Hosono (Tokyo Institute of Technology, Yokohama, Japan); G. Chen, W. Yu (Renmin University of China, Beijing, China); and B. Moeckly (STI, Santa Barbara, CA)

The behavior of vortex matter in superconductors arises from the competition of three characteristic energies, namely those resulting from inter-vortex interactions, thermal fluctuations, and interactions of vortices with the inhomogeneities in the material (defects). If all these ingredients are appropriately accounted for, it should be possible to develop a universal description of vortex matter, applicable to all superconductors. The present understanding of vortex physics falls far short of that expectation, mostly because of the complexity of the interactions of vortices with inhomogeneities. Our goal is to develop a general quantitative description of vortex matter in superconductors, and our approach is to compare and contrast systems with vastly different superconducting properties under a broad spectrum of conditions including extreme ones.

In this poster we will present a comparative analysis of the vortex physics in high temperature (or oxide) superconductors (HTS), iron-arsenides, and MgB_2. The oxide HTS exhibit a rich vortex phenomenology due to the strong influence of thermal fluctuations, which is a consequence of the small superconducting coherence length (ξ) and the large crystalline anisotropy (γ). Iron-arsenide superconductors represent a whole new family of materials spanning broad ranges of T_c, ξ and γ, where the generality of the ideas initially developed for HTS materials can be tested. In addition, the multi-band superconductivity in the FeAs-based compounds introduces a new level of complexity. Valuable information can also be obtained from studies in MgB_2, a chemically simpler two-band superconductor where ξ and γ can be modified by doping.

Ions to wires: The science of high Tc superconducting wires

T.G. Holesinger (MPA-STC)

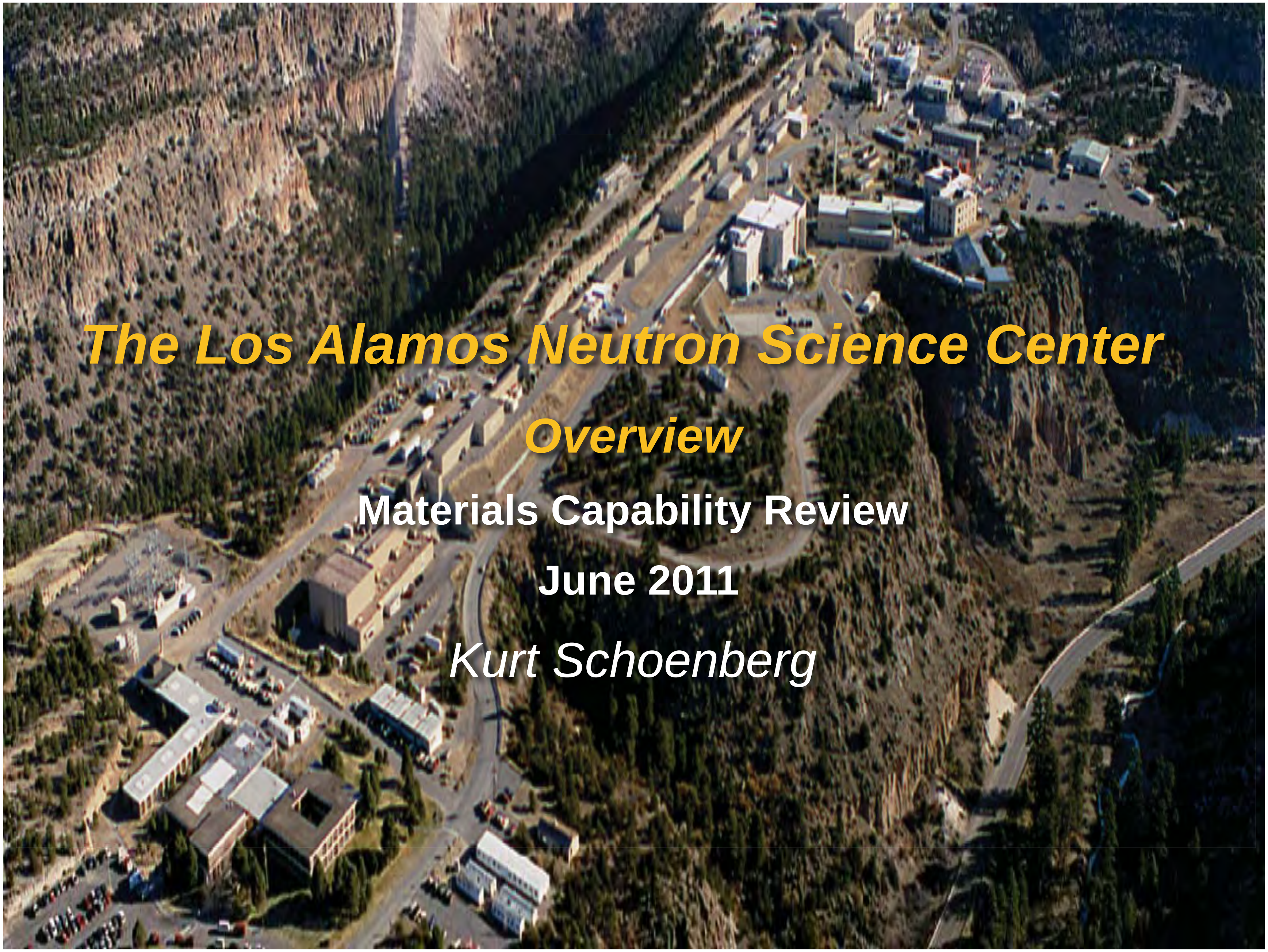
Important aspects of LANL's material capability include science guiding discovery and the transformational science of discoveries into devices. The superconductivity program in MPA-STC is a success in both respects. Los Alamos National Laboratory (LANL) is assisting the Department of Energy Office of Electricity Delivery and Energy Reliability (OE) to lead national efforts to modernize the electric grid, enhance security and reliability of the energy infrastructure, and facilitate recovery from disruptions to energy supplies. This program works closely and in partnership with U.S. manufacturers, utilities, project developers, federal and state policy makers, and end-users to advance technology, reduce development and operating costs to ensure a more reliable and secure electric and energy infrastructure. LANL pioneered the development of the $YBa_2Cu_3O_y$ (YBCO) coated conductor technology transferred to industry in this country and adopted around the world. Included were the development of film processes for textured, single-crystal like templates that can be manufactured on the kilometer length scale and superconducting films with nano-engineered microstructures and tunable properties. Advanced characterization processes were developed for linking structure-property-chemistry relationships, understanding optimization processes in wire development, and minimizing AC losses in fully processed wires. LANL continues to define extreme performance in coated conductor materials, establishing world records for performance in self and applied magnetic fields. Critical current densities of 5 MA/cm^2 have been measured in 2 μm thick YBCO films (critical currents of 1000 A/cm-width) with self-assembled $BaZrO_3$ nanorods and Y_2O_3 nanoparticle layers. Flux pinning forces of 32 and 122 GN/m^3 have been measured at 75 and 65 K, respectively, in YBCO films with self-assembled YBa_2NbO_y nanorod structures. LANL maintains a number of ongoing industrial interactions for transferring technology and supporting American industry's leadership in this area. LANL's longest, continuous industrial partnership is, in fact, a collaboration of 23 years with American Superconductor.

The Los Alamos Neutron Science Center: Status and future plans

K. Schoenberg (ADEPS)

The Los Alamos Neutron Science Center (LANSCE) has served the international research community for more than 30 years as a premier facility for fundamental and applied science. The heart of the LANSCE facility is the 800-MeV linear accelerator system (LINAC) that is capable of accelerating both positive and negative hydrogen ions and of delivering those beams to multiple experimental areas simultaneously. Three experimental areas, the Manuel Lujan Jr. Neutron Scattering Center (Lujan Center), the Weapons Neutron Research (WNR) Facility, and the Proton Radiography Facility (pRad), are designated DOE national user facilities. In addition, a the Ultra Cold Neutron research facility, that explores fundamental nuclear physics, and the Isotope Production Facility (IPF), that produces a wide range of radioisotopes for medical diagnosis, treatment, and scientific research, comprise the balance of LANSCE research facilities.

Future scientific missions will require enhanced LANSCE capabilities to support five principal research areas: 1) condensed matter science, 2) fundamental nuclear physics, 3) applied nuclear science and technology, 4) bioscience, and 5) materials under extreme conditions. The LANSCE enhancement strategy is composed of two parts; enhancements to LANSCE facilities that fully exploit existing capabilities using 800 MeV protons, and investment in the LINAC and beam transport systems that will enable significant improvement to facility reliability and performance. Central to this enhancement strategy is the LANSCE LINAC risk mitigation project that was started in 2010. This talk will summarize the status of present LANSCE facilities and research as well as future plans.



The Los Alamos Neutron Science Center

Overview

Materials Capability Review

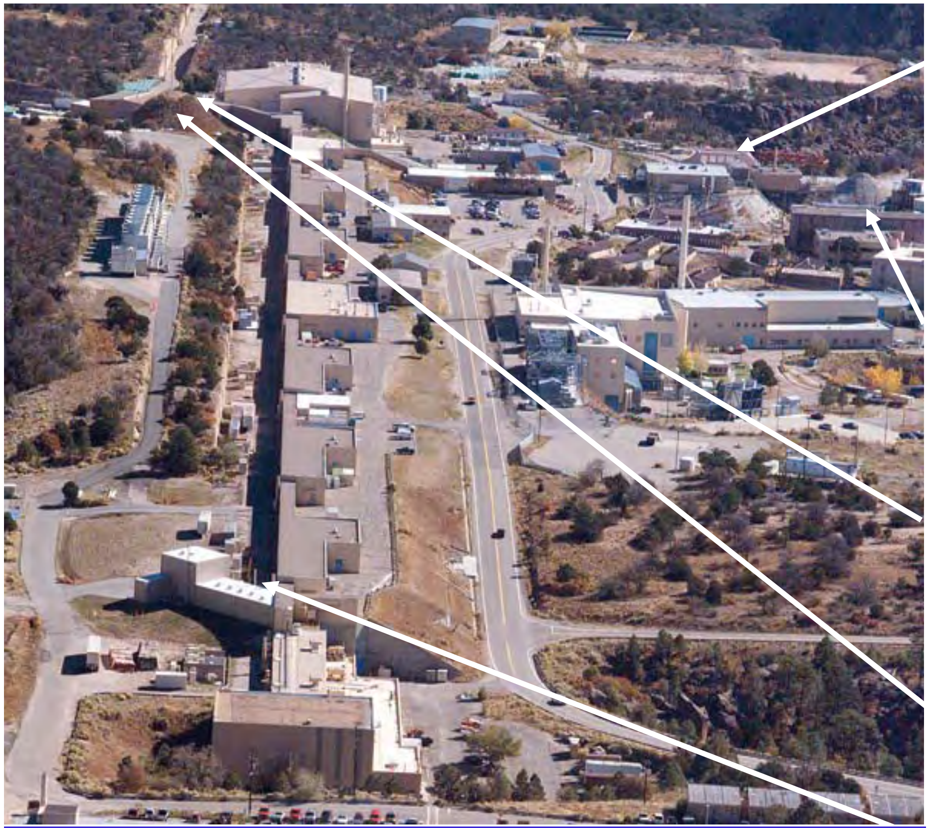
June 2011

Kurt Schoenberg

Outline

- ***LANSCE Today***
- ***Our performance***
- ***LANSCE Tomorrow***
  - ***Risk Mitigation (LRM)***
  - ***Future Initiatives***

LANSCE presently provides the US and international research communities a diverse set of premier facilities



Unique, highly-flexible beam delivery to multiple facilities 6 mo/yr @ 24/7 with ~ 1200 user visits

Lujan Center

- ***Materials science and condensed matter research***
- ***Bio-science***
- ***Nuclear physics***
- ***A National BES user facility***

WNR

- ***Nuclear physics***
- ***Semiconductor irradiation***

Ultra-cold Neutron Facility

- ***Fundamental nuclear physics***

Proton Radiography

- ***HE science, dynamic materials science, hydrodynamics***

Isotope Production Facility

- ***Nuclear medicine***
- ***Research isotope production***

LANSCE facilities support principal Laboratory missions and research needs

Research area	Needs/Drivers	Thrusts	Facility
Materials and Bioscience	National Security Materials science Bioscience Ties to materials strategy	Processing-structure-performance Fundamental properties Short/long range order Processing-structure-performance Superconductivity, Hydrogen storage…. Biotoxin mechanisms Protein function (location of Hydrogen) Self-assembly -Emergent Phenomena -Defects and Interfaces	Lujan
Nuclear Science	National Security Nuclear energy Astrophysics Other nuclear physics	Precision Fission, outputs, capture: materials and diagnostics Fission, capture: advanced fuels Capture, nucleosynthesis processes Level densities	WNR Lujan
Materials Dynamics	National Security Ties to materials strategy	High explosives, shock dynamics, material damage, implosion dynamics - Extreme Environments	pRad
Extreme Radiation Environments	National Security Advanced fuels Semiconductor upset Ties to materials strategy	Weapon component qualification High power fuel irradiation testing Industry standard for testing, cosmic ray upset - Extreme Environments	WNR MTS WNR
Fundamental Nuclear Science	Particle properties Beyond standard model research	Ultracold neutron collaboration	UCN
Isotopes	Medical therapy Medical, Physics Environment National Security	Production for National Clinical Use (Sr^{82}, Ge) Short lived isotopes Short lived isotopes – SSMP, attribution	IPF

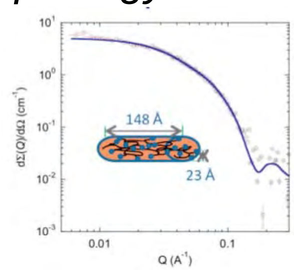
LANSCE Research Examples: MCR 2011 Posters

- **Radiation Environments**
  - *Irradiation Environment in the Proposed Materials Test Station (E. Pitcher)*
  - *Nuclear Physics and Material Science (J. Ullmann)*
- **Nuclear Energy – Actinide Focus**
  - *Residual Stresses in Mini Monolithic U10Mo Fuel Plates (D. Brown)*
  - *Chemical Segregation of U-10 Mo Fuel Foils During Simulated Bonding (S. Vogel)*

    Neutron diffraction provides a unique capability to characterize phase transformation kinetics, chemistry, texture, and strains during iso-thermal annealing of metallic nuclear fuels.

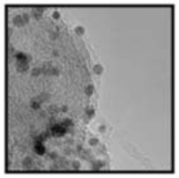
- **Materials for Clean Energy**
  - *Connecting Ionomer Dispersion Morphology to Fuel Cell (C. Welch)*

Neutron Connecting the dots: Relationship between dispersion and film morphology

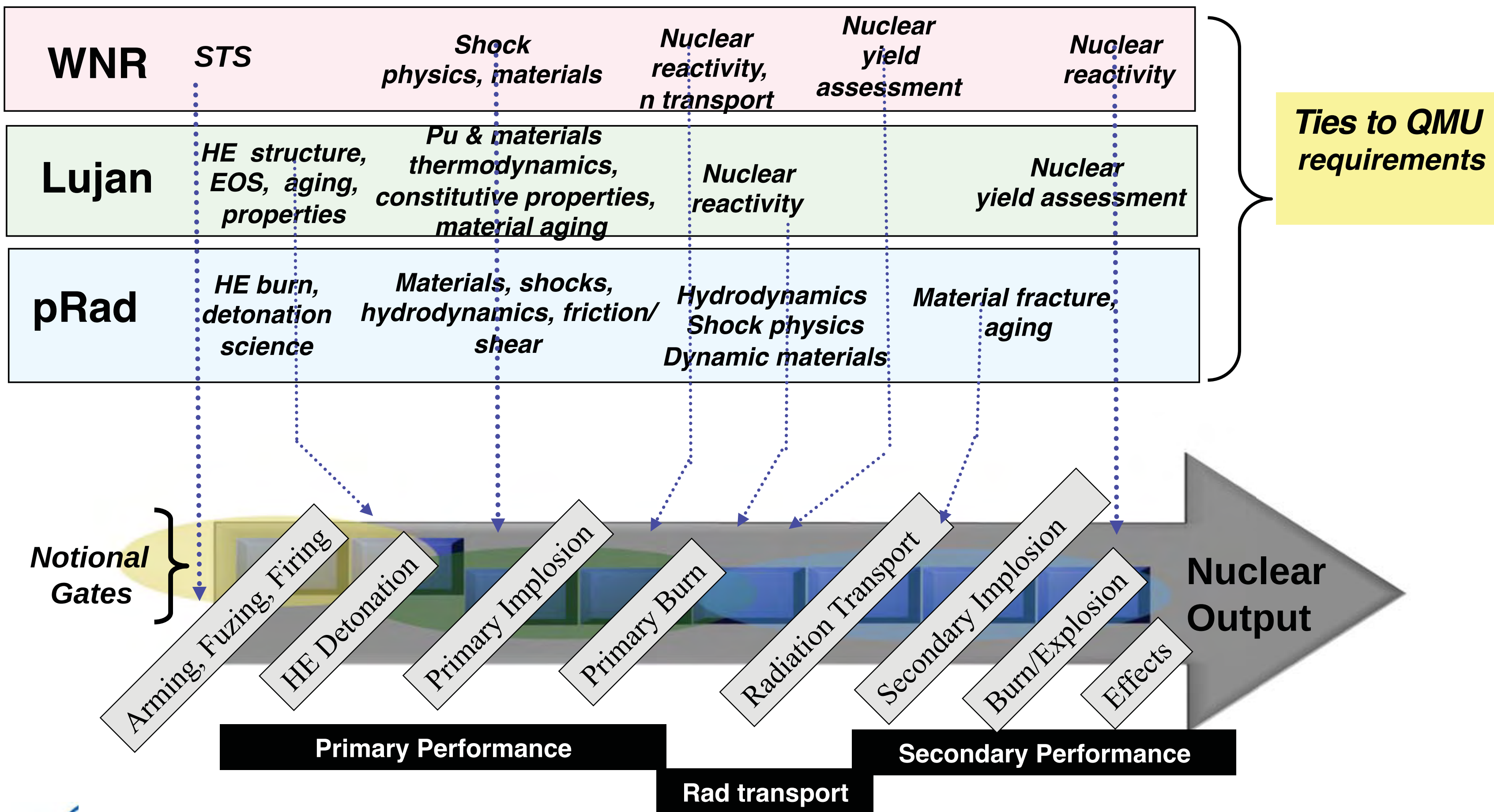


Polymer Morphology Dispersion

Electrode Morphology

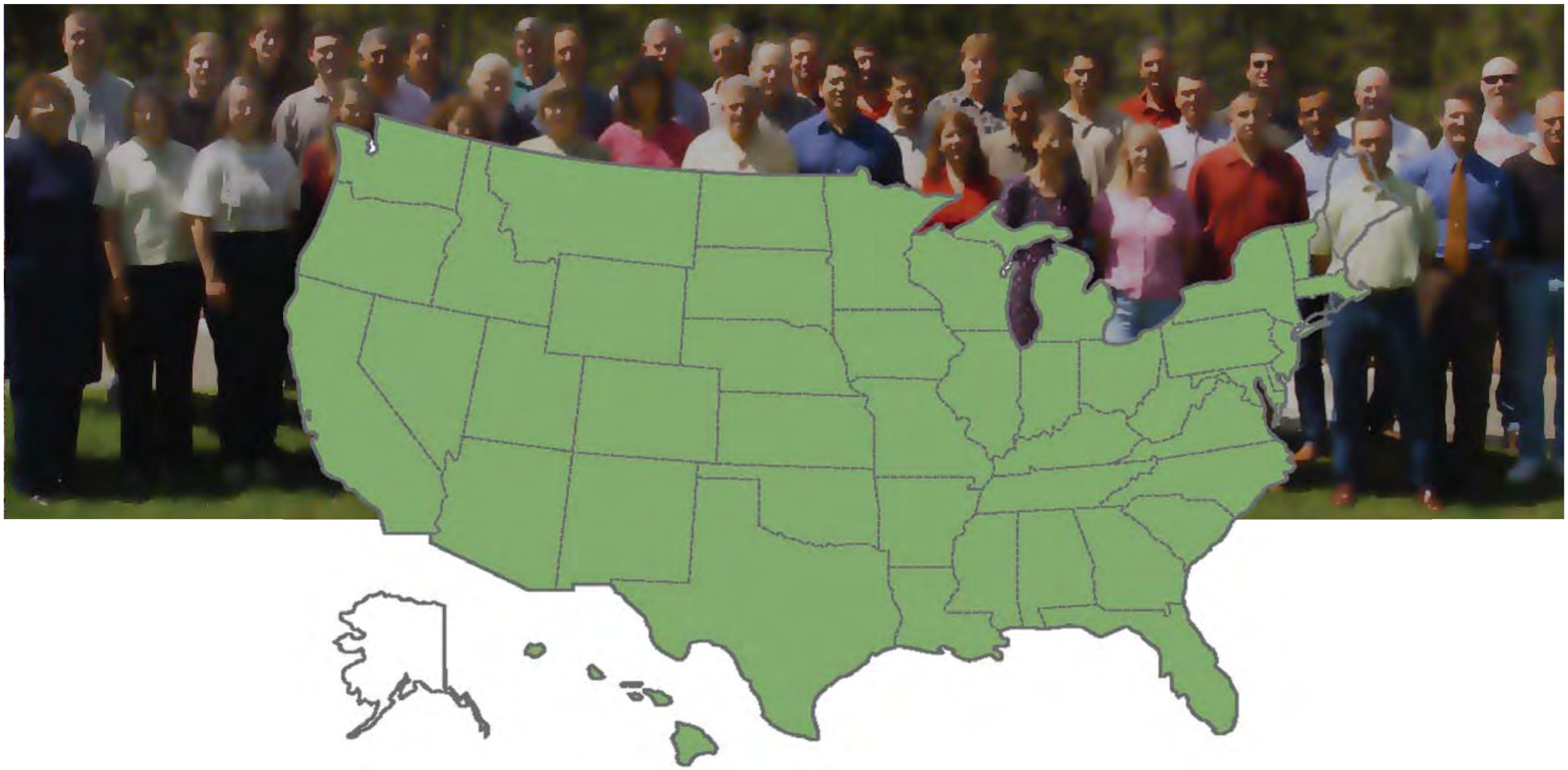


LANSCE facilities are unique and support developing the broad science-based predictive capabilities required for future certification



LANSCE facilities address a broad range of DP research requirements

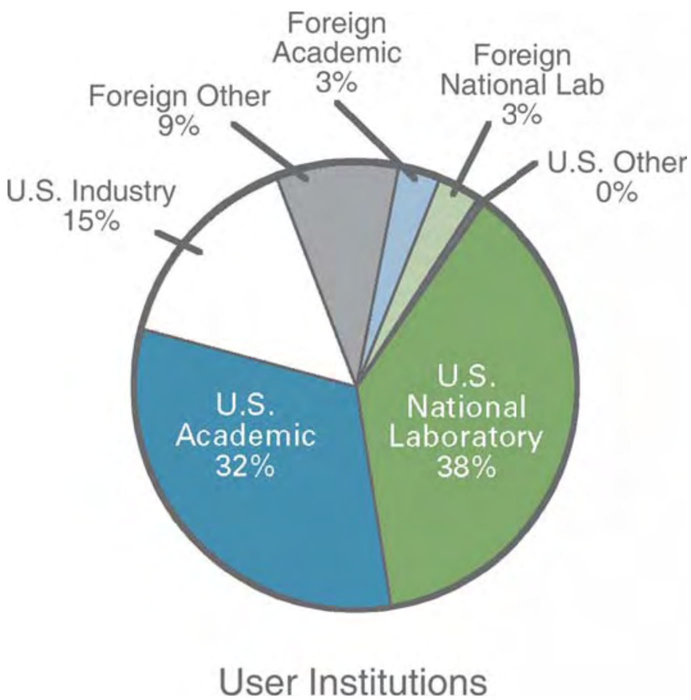
Lujan, WNR, and pRad are designated DOE National User Facilities with a Nationwide clientele

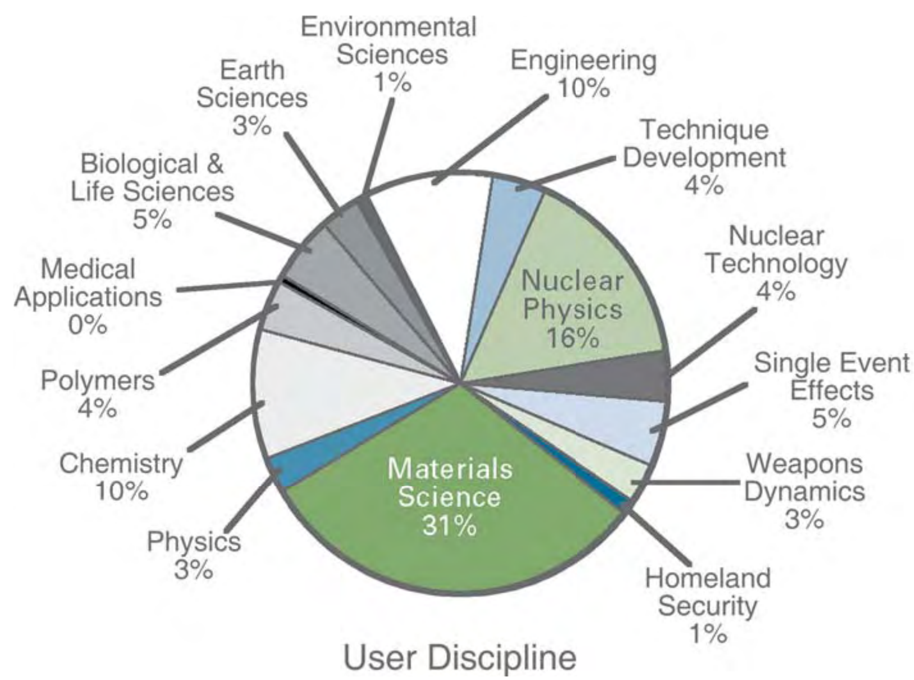


■ U.S. States Represented by LANSCE Users

LANSCE users comprise a diverse and evolving user community

Present LANSCE: 1200 User Visits Annually: 45 states, 15 foreign countries





Outline

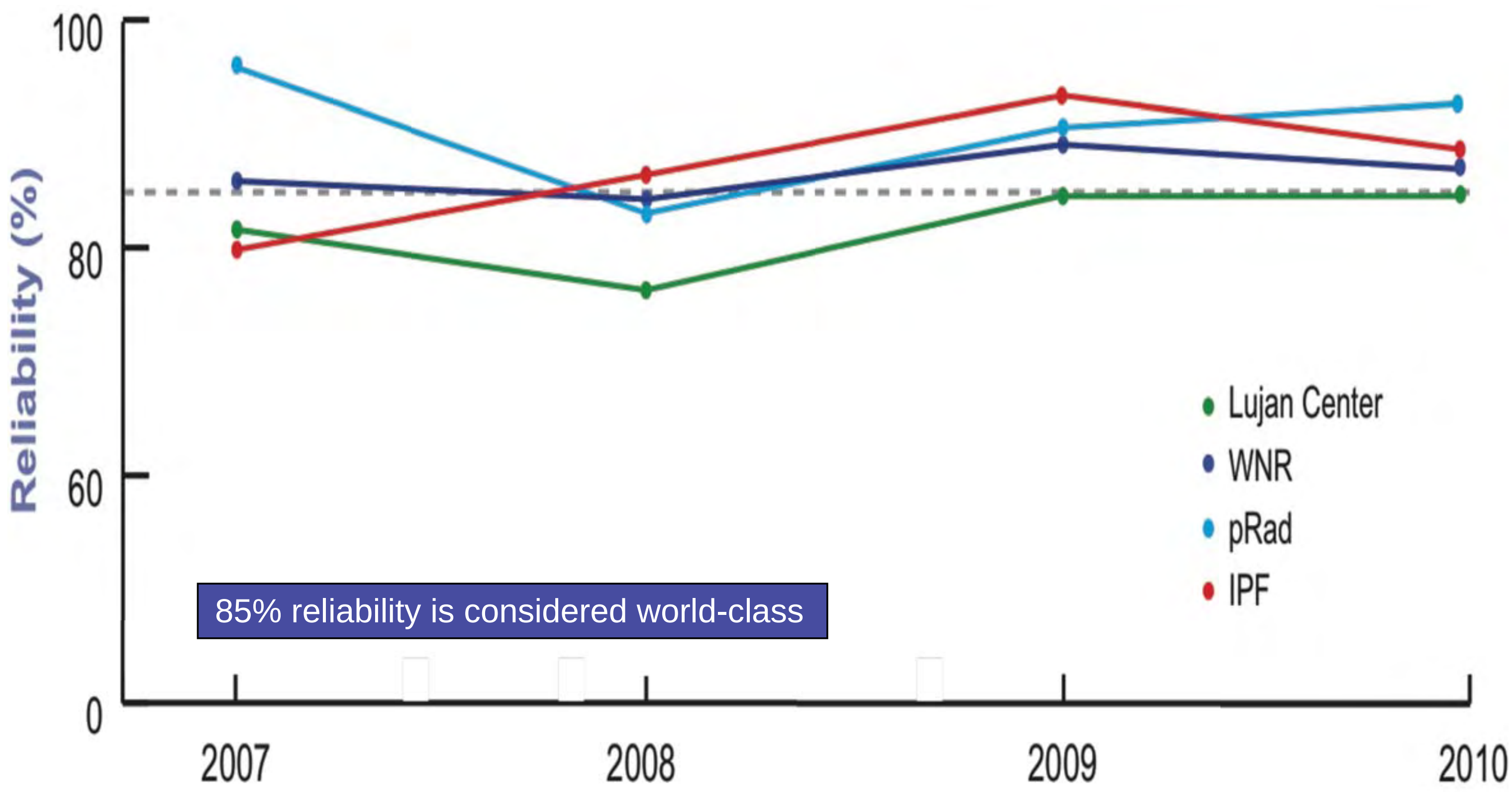
- *LANSCE Today*
- ***Our performance***
- *LANSCE Tomorrow*
    - *Risk Mitigation (LRM)*
    - *Future Initiatives*

LANSCE Calendar Year 2010 beam reliability was excellent

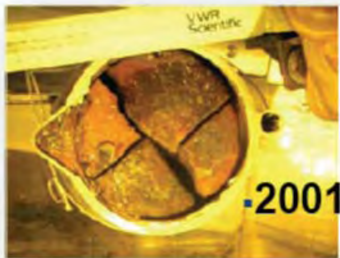
Target	Hours Scheduled	Hours Delivered	Reliability
Lujan Center	3390	2868	84.6%
WNR Target 4	3041	2635	86.7%
pRad	821	766	93.4%
IPF	4165	3660	87.9%
UCN	1800	1625	90.3%
WNR Target 2	499	441	88.4%

- The LINAC is operating with over 90% reliability

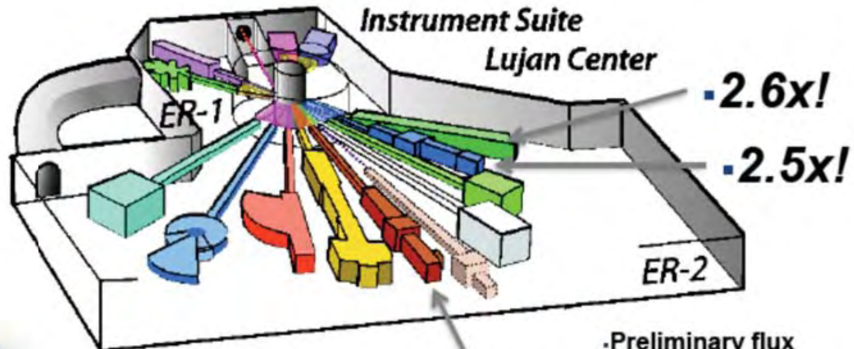
LANSCE has demonstrated world-class operational reliability for all facilities over the past 2 years



The new Mark III target delivers 2X to 3X more flux through design innovation – intrinsic brightness equivalent to SNS

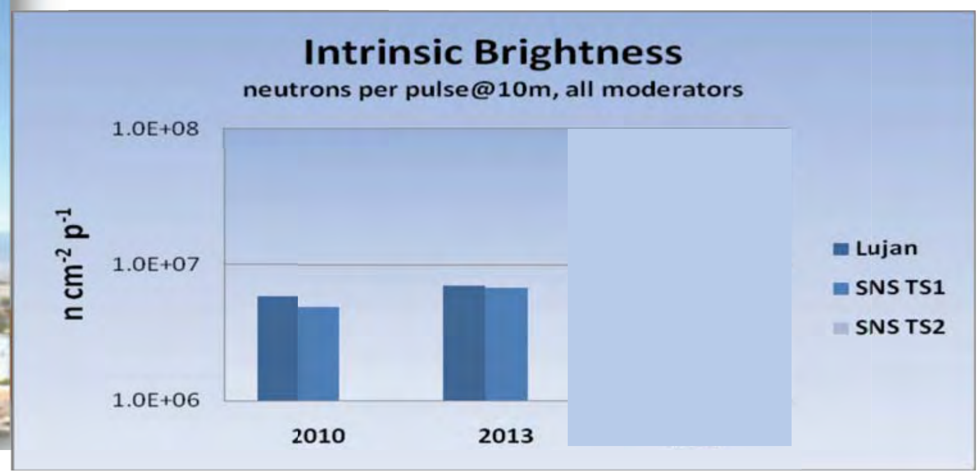


- New Ta cladding will minimize corrosion

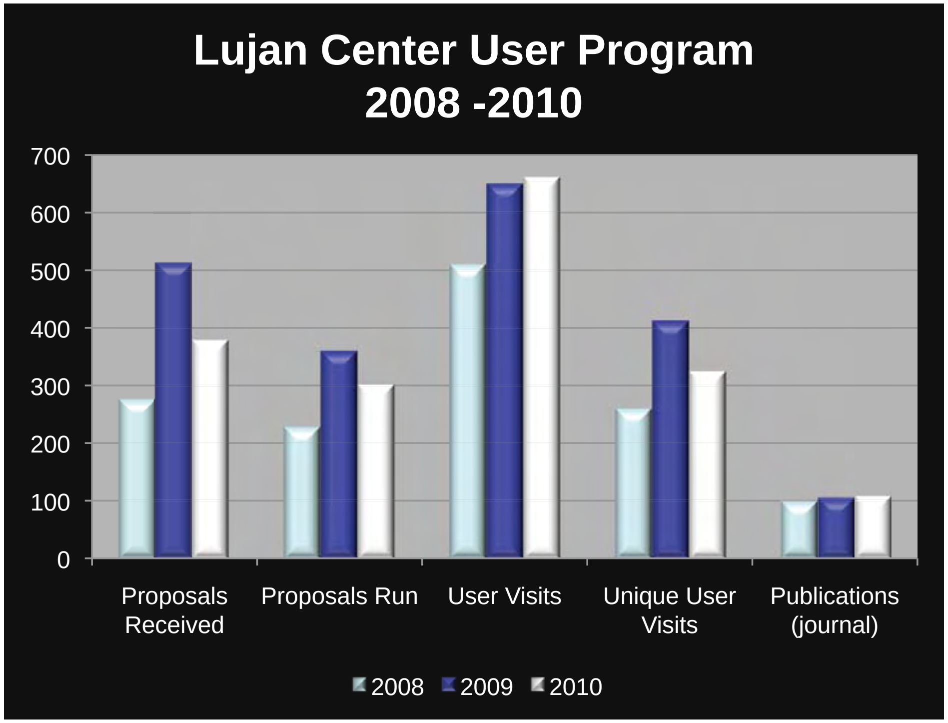


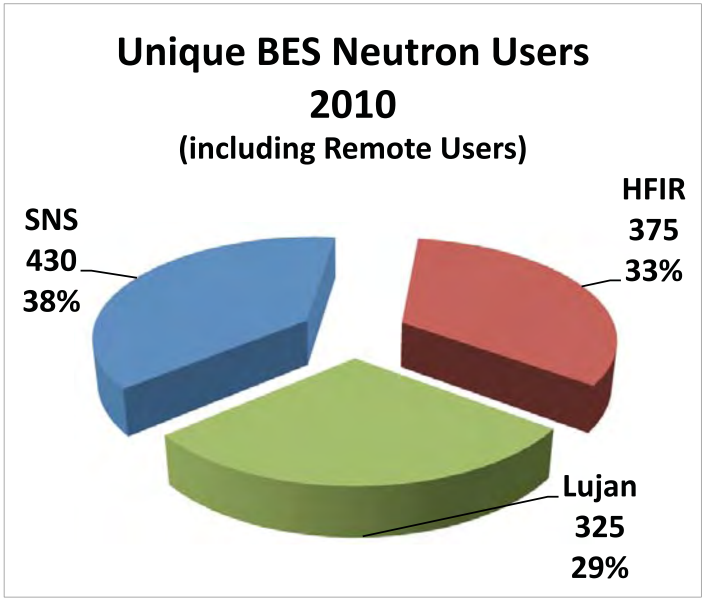
- Preliminary flux measurements indicate increases on all beamlines

- New Be reflector filter increases cold neutron flux



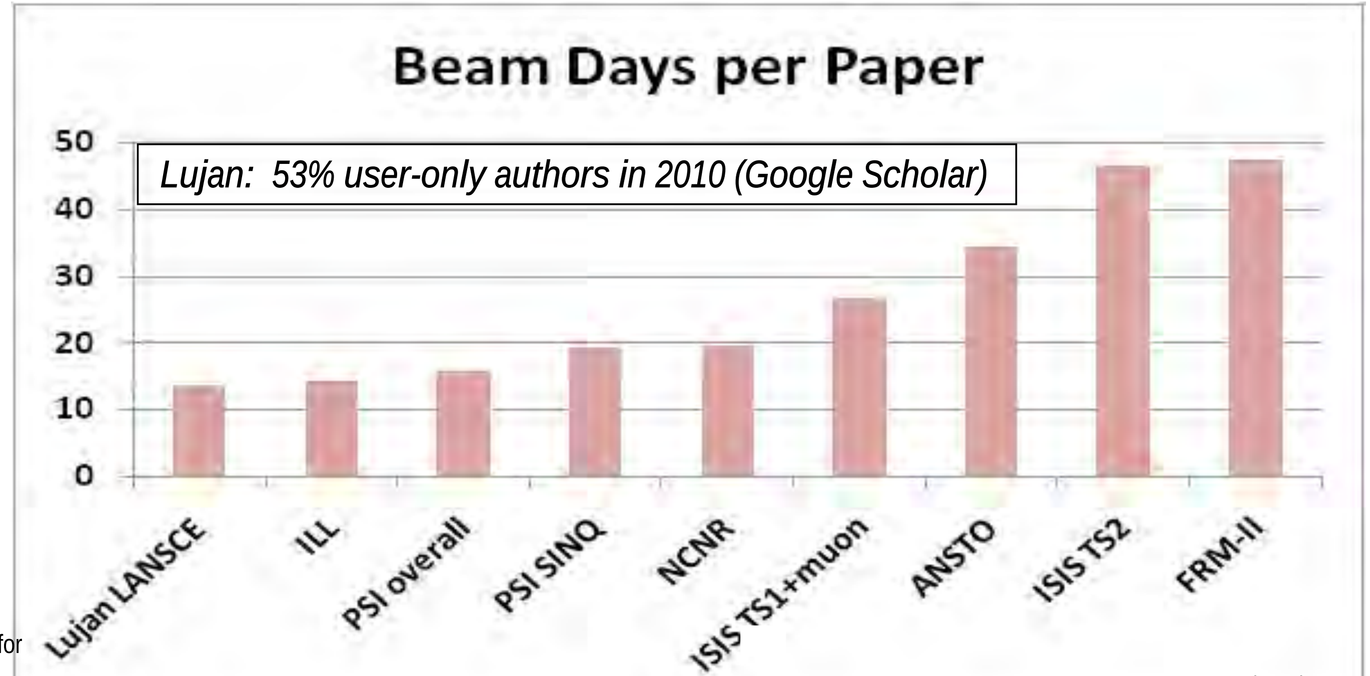
Los Alamos Neutron Science Center
Lujan Center Statistics





DOE Neutron Centers: LANSCE-Lujan is a major player, delivering science and contributing to the growth of the US Neutron Community

Lujan Center users enjoy an outstanding environment for publishing



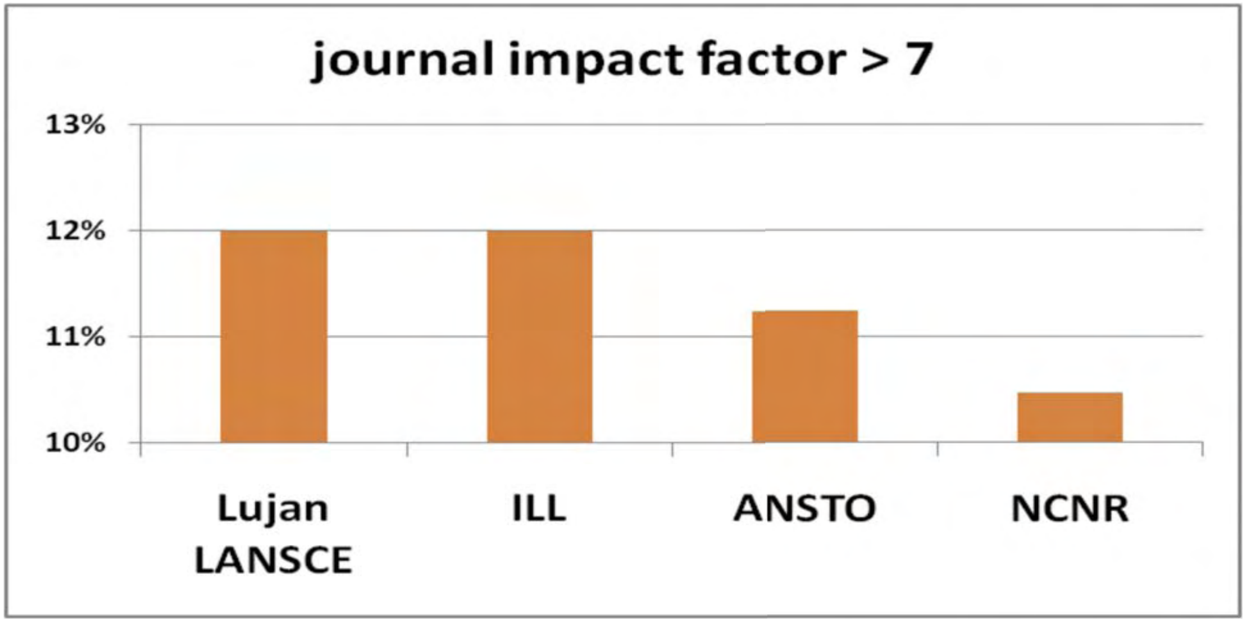
- Sources for
  - (n,d,p)

(1,4,3) (1,1,2) (1,2,2) (1,2,2) (2,2,2) (1,2,2) (4,4,4) (1,2,2) (1,1,2)

- n=number of instruments listed on web as active, including radiography, nuclear physics and muon science when applicable
- d=number of days of operations
- p=number of archival journal publications, including radiography, nuclear physics and muon science when applicable

1. web for 2010
2. 2010 activity reports, on web or hard copy
3. 2010 Google Scholar search
4. personal communication

Lujan Center high-impact publication fraction is competitive with the best neutron scattering laboratories



- Impact factor taken from 2008 ISI report
- ILL data taken from "Vettier Report" 2005
- ANSTO data from personal communication (R. Robinson)
- NCNR data taken from 2010 Activity Report
- Lujan data taken from Google Scholar

Outline

- *LANSCE Today*
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The three Undersecretaries of Energy recently reaffirmed the importance of LANSCE to the Laboratory and the Department

LANSCE is currently performing programmatic research of importance to the National Nuclear Security Administration as well as fundamental science for the Office of Science and materials research and testing relevant to nuclear energy.

We anticipate that the important research being done now will continue for at least a decade and expect LANL to maintain the capabilities of LANSCE through an enhanced maintenance program … to ensure LANSCE is available over this time period.

Department of Energy
Washington, DC 20585

March 24, 2010

Dr. Michael Anastasio
Director
Los Alamos National Laboratory
P. O. Box 1663
Los Alamos, NM 87545

Dear Dr. Anastasio:

We are writing to clarify the Department's expectations for the future of the Los Alamos Neutron Science Center (LANSCE). LANSCE is currently performing programmatic research of importance to the National Nuclear Security Administration as well as fundamental science for the Office of Science (including the Lujan Center Neutron Scattering Facility for Basic Energy Sciences and the Isotope Production Facility for Nuclear Physics) and materials research and testing relevant to nuclear energy. We further understand that some important maintenance activities for the accelerator that is the heart of LANSCE have been deferred while the LANSCE-Refurbishment (LANSCE-R) project was being formulated. The recent termination of the LANSCE-R project has led to some concerns regarding the future of the LANSCE facility that we would like to address.

Specifically, we anticipate that the important research being done now will continue for at least a decade and expect Los Alamos National Laboratory (LANL) to maintain the capabilities of LANSCE through an enhanced maintenance program, including those maintenance activities deferred while the LANSCE-R project was being initiated. We are requesting that LANL provide a plan to us regarding the full suite of maintenance issues that need to be addressed in order to sustain today's level of operations through the decade. This plan should include the maintenance to ensure that LANSCE is available over this time period.

In the longer term, we would like to continue to engage you in a discussion of the need for high quality experimental science capabilities at LANL to ensure the intellectual vitality of the Laboratory and ensure its continued ability to support important

LANSCE Risk Mitigation Strategy is discussed in the Laboratory Director's response to the 3 Undersecretaries letter

Our plan is three fold:

1)Increase the annual operating budget to a level that supports execution of the essential maintenance to allow the linac to operate in the short-term

2)Invest in long-term capital replacements to mitigate the major risks to continued operation of the linac and beam transport systems

3) Invest in risk mitigation for the rest of the facility to provide for a more reliable capability

Los Alamos
NATIONAL LABORATORY
EST. 1943

Office of the Director

July 26, 2010

Tom D'Agostino
U.S. Department of Energy
Forrestal Building
1000 Independence Ave SW
Washington, D.C. 20585

Kristina Johnson
U.S. Department of Energy
Forrestal Building
1000 Independence Ave SW
Washington, D.C. 20585

Steven E. Koonin
U.S. Department of Energy
Forrestal Building
1000 Independence Ave SW
Washington, D.C. 20585

Dear Under Secretaries D'Agostino, Johnson, and Koonin:

On March 24, 2010, I received your letter articulating the Department's expectations for the future of the Los Alamos Neutron Science Center (LANSCE). I am gratified at the recognition of the broad impact that LANSCE has on the mission of Los Alamos National Laboratory. LANSCE is the premier experimental facility at the Laboratory, and used by many LANL projects and hundreds of international users annually.

In your letter, Los Alamos was requested to provide a plan regarding the full suite of issues that need to be addressed in order to sustain today's level of operations through the decade. You also asked that we address the longer-term need for high quality experimental science capabilities, and suggested materials under extreme conditions as a potential focus area. I appreciate your recognition of the importance of LANSCE in the near term, and the importance of a signature experimental capability for the longer term. I address both questions in this letter and its enclosures.

Our plan for LANSCE is three-fold: increase the annual operating budget to a level that supports execution of the essential maintenance that continues to allow the linac to operate in the short term; invest in long term capital replacements to mitigate the major risks to continued operation of the linac and beam transport systems; and invest in risk mitigation for the rest of the facility to provide for a more reliable capability. This plan entails use of the prior year's LANSCE-R funding to address the highest risk elements and a subsequent investment of approximately $25M per year for 5 years to address the first two elements. The details of these three elements are described in the enclosure entitled *LANSCE Linac Risk Mitigation Strategy*. I appreciate the help that you have provided in re-programming the LANSCE-R money for this purpose. We have taken action to identify the needed operational budget for LANSCE, and now I am asking your help to provide $25M (through Defense Programs) in the FY12 budget to continue the linac risk mitigation effort.

We have been studying the issue of long-term science facility development for approximately five years. We have concluded that Matter-Radiation Interactions in Extremes (MaRIE), is the appropriate signature experimental facility for LANL. MaRIE will revolutionize materials in extremes, a key challenge that spans a broad range of DOE missions, through unprecedented *in situ*, transient

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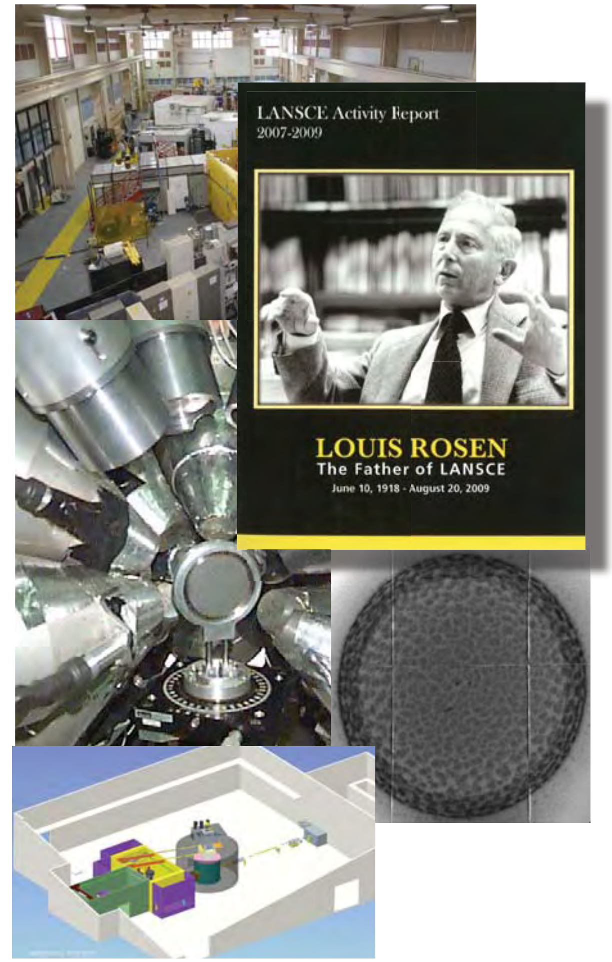
LINAC risk mitigation investments ensure that LANSCE reliably operates to meet our national research needs well into the 21st century

The LANSCE LINAC

- **LINAC risk mitigation investments are designed to:**
  - ***Refurbish the 201MHz and 805 MHz RF systems to regain reliable RF power system operation.***
  - ***Restore 120 Hz linac operation.***
  - ***Implement a modern, maintainable EPICS-based control system.***
  - ***Refurbish beam transport and front-end injector systems (RFQs)***
- **Work is being integrated with operations to ensure continued programmatic research and a robust user program during project execution.**

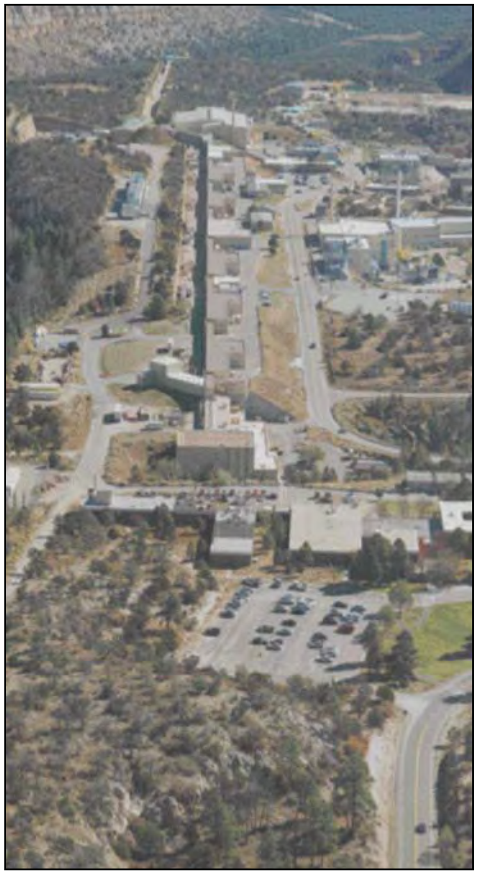
LANSCE 2015 on the path to MaRIE: Continue to enhance our capabilities at 800 MeV to meet future science and technology missions

- **Material Science: The Lujan Enhancement Project will fully enhance capabilities at Lujan in cold neutron scattering with extreme environments.**
- **Dynamic Materials Research: Explore materials properties and hydrodynamics of dynamic and scaled systems using evolving pRad capabilities**
- **Nuclear Science: "Pulse-stacking" in the PSR will greatly enhance research on unstable isotopes and will expand unique WNR capabilities (eV to MeV).**
- **Nuclear Technology at Extremes: Materials irradiation and isotope production at the Materials Test Station and improved semiconductor irradiation at WNR**



Conclusion: LANSCE will continue to play an important role in providing capabilities for key fundamental and programmatic science

- **Capabilities support, and are adapted to, Lab national security and science missions.**
- **Interplay of basic and national security missions is unique and provides unique opportunities for innovation in basic and applied science.**
- **The LANSCE User Facility is operating better than ever, supporting a broad range of fundamental and applied science based on three neutron sources, and producing medical radioisotopes for diagnostic imaging.**
- **The scientific portfolio is robust and diverse, utilizing neutrons that vary in energy by fifteen orders of magnitude.**
- **LINAC risk mitigation investments are moving us to future reliable operations.**
- **The LANSCE facility will form the foundation for MaRIE, the future signature science facility at LANL**




Welcome to LANSCE

Materials Review Committee

GARY S. WAS (Chair)

Department of Materials Science and Engineering
Department of Nuclear Engineering and Radiological Sciences
Director, Michigan Memorial Phoenix Energy Institute
University of Michigan

Gary S. Was received his Sc.D. from MIT in 1980. He is the Walter J. Weber, Jr. Professor of Sustainable Energy, Environmental and Earth Systems Engineering, and holds appointments in Nuclear Engineering and Radiological Sciences, and Materials Science and Engineering at the University of Michigan. He has held positions as director of the Michigan Memorial Phoenix Energy Institute, associate dean of the College of Engineering and chair of the Nuclear Engineering and Radiological Sciences Department. Professor Was's research is focused on materials for advanced nuclear energy systems and radiation materials science, including environmental effects on materials, radiation effects, ion beam surface modification of materials and nuclear fuels. He has worked extensively in experiments and modeling of the effects of irradiation, corrosion, stress corrosion cracking and hydrogen embrittlement on iron- and nickel-base austenitic alloys. Was has led the refinement of models for radiation induced segregation to account for composition dependent processes, and developed the first comprehensive thermodynamic and kinetic model for chromium carbide formation and chromium depletion in nickel-base alloys. Most recently his group has led the development of proton irradiation as a technique for emulating neutron irradiation effects in reactor structural materials and has conducted some of the first stress corrosion cracking experiments of austenitic and ferritic alloys in supercritical water. He has published more than 340 papers in archival journals or refereed conference proceedings, and a graduate textbook on radiation materials science. He is a Fellow of the American Nuclear Society, ASM International, NACE International, and the Materials Research Society.

THOMAS B. BRILL

Chemistry and Biochemistry
University of Delaware

Thomas Brill is a professor of chemical engineering at the University of Delaware. He received his B.S. (1966) with high honors in chemistry from the University of Montana and his Ph.D. (1970) in inorganic chemistry from the University of Minnesota. He has taught at the University of Montana, the University of Minnesota, and North Carolina State University. He has been an assistant, associate, and full professor at the University of Delaware. He spent two years as a visiting professor at the University of Oregon and at Zhongshan University in the Peoples' Republic of China.

Professor Brill has lectured widely at numerous universities and conferences in the United States and abroad dealing with such topics as energetic materials, molecular processes in condensed matter, advanced oxidation technologies, and emerging technologies in hazardous waste management. He has been a consultant to business, industry, and national laboratories. His current research topics are structure and decomposition mechanisms of energy-rich compounds; chemistry of a burning surface; reactions in supercritical water; solid-state chemical processes; Fourier-transform infrared spectroscopy; laser Raman spectroscopy; synthesis, structure, and bonding of inorganic and organometallic complexes; and the Arbuzov reaction.

He has published 248 research papers and 3 books and has given 212 conference presentations. He is a member of professional organizations including the American Chemical Society and the Materials Research Society and was named Fellow of the Center for Advanced Studies. He is listed in such publications as *Who's Who in Science and Technology* and *Who's Who in the United States*.

BARBARA JONES

IBM Research - Almaden

Barbara Jones received her Ph.D. in physics from Cornell University in 1988. She is the group leader of Theoretical and Computational Physics at IBM Almaden Research Center. Her research interests include analytic and computational studies of strongly correlated electrons and quantum magnetism especially at the atomic scale, current-induced magnetization reversal and spin torque, Griffiths phase and disorder effects in heavy fermions, and the thermodynamics of viral evolution. Jones has served on several committees including the National Academies Solid State Sciences as Chair, *Physical Review B* Editorial Board, and the Argonne Center for Nanoscale Materials Advisory Board.

M. BRIAN MAPLE
Department of Physics
University of California, San Diego

M. Brian Maple is a professor in the Department of Physics at the University of California, San Diego. He received his Ph.D. from UC, San Diego in physics in 1969. He is the Director for the Institute for Pure and Applied Physical Sciences and the Center for Interface and Materials Science, both at the university. Maple is presently the Bernd T. Matthias Endowed Chair in Physics. He has served on several national laboratory committees, is a National Academy of Sciences member, an American Physical Society Fellow, an American Association for the Advancement of Science Fellow, a Materials Research Society member, and serves on many other committees for the materials community.

I. CEVDET NOYAN
Department of Applied Physics & Applied Mathematics
Columbia University

I. Cevdet Noyan is a professor in materials science and engineering at Columbia University. He received his Ph.D. from Northwestern University in materials science and engineering in 1984. Dr. Noyan is a Fellow in the American Physical Society, is the U.S. representative to the International Residual Stress Analysis Conferences, a member of ASM, TMS-AIME, ACA, APS, Sigma Xi, and IEEE. He holds 23 patents. Noyan's research interests lie in mechanical response of crystalline materials over various length scales. He was one of the first researchers to combine the theory of micromechanics with that of x-ray and neutron diffraction. From 2005 on, he was a co-principal investigator on the new X13B microbeam diffraction line was built at NSLS and commission in 2008.

RALPH NUZZO
Department of Chemistry
University of Illinois at Urbana-Champaign

Ralph G. Nuzzo is the William H. and Janet G. Lycan Professor of Chemistry at the University of Illinois at Urbana-Champaign, a faculty he joined in 1991 and where he also holds an appointment as a professor of materials science and engineering. He received an A.B. degree with high honors and highest distinction in chemistry from Rutgers College in 1976, where he was also recognized as a Henry Rutgers Scholar, awarded the Merck Prize for undergraduate research, and elected to Phi Beta Kappa. He earned a Ph.D. in organic chemistry from The Massachusetts Institute of Technology in 1980. He accepted the position of member of technical staff in materials research at Bell Laboratories in Murray Hill, NJ in 1980, where he was named a distinguished member of the staff in research in 1987—a title held until he left to join the Illinois faculty in 1991.

Professor Nuzzo is a Fellow of the American Academy of Arts and Sciences and Fellow of the World Innovation Foundation. In 2006 he was recognized by a *Wall Street Journal* Innovators Award for Semiconductors. He received the Adamson Award of the American Chemical Society in 2003 for original discoveries leading to the development of self-assembled monolayers. Nuzzo has been recognized three times by ISI for Citations in Chemistry, and the American Chemical Society—*Journal of the American Chemical Society*—for his co-authorship of one of the 12 most highly cited papers published in the journal during its 125 year history.

Nuzzo is the senior editor of *Langmuir* and serves on the advisory, review, and executive boards of numerous entities—ones both public and private. He is a senior advisor to the Dreyfus Foundation and the Petroleum Research Fund, and a member of the scientific advisory boards of Surface Logix Inc. and the Intermolecular Corporation. He more recently co-founded and serves as a member of the board of Directors of the Semprius Corporation. Nuzzo holds 16 U.S. patents, and is the author of more than 200 primary research publications.

ADAM SCHWARTZ
Condensed Matter and Materials Division
Lawrence Livermore National Laboratory

Adam Schwartz is the division leader for Condensed Matter and Materials Division in the Physical and Life Sciences Directorate at the Lawrence Livermore National Laboratory (LLNL) in Livermore, California. His research focuses on the fundamentals of plutonium aging, phase transformations and phase stability of plutonium alloys, electronic structure of actinide elements, and dynamic properties of materials. These research efforts are supported by the National Nuclear Security Administration. He holds B.S. and M.S. degrees in metallurgical engineering from University of Pittsburgh, and a Ph.D. degree in materials science and engineering from the University of Pittsburgh. In addition to serving as the division leader, he is the deputy program leader for dynamic materials properties within the science campaigns and the deputy program leader for physics and engineering models in the Advanced Simulation and Computing program. Prior to these activities, he led the Plutonium Aging and Pit Lifetime program at LLNL, was major technical effort leader for Pu aging, was principal investigator on numerous projects, and was a postdoctoral research associate at LLNL.

SARAH TOLBERT
Department of Chemistry & Biochemistry
University of California, Los Angeles

Sarah Tolbert received her B.S. summa cum laude in chemistry from Yale University in 1990 and a Ph.D. in materials physical chemistry from the University of California, Berkeley in 1995, working with professor Paul Alivisatos. After graduating, she went to the University of California, Santa Barbara as an NSF Postdoctoral Fellow working with professor Galen Stucky. She joined the Department of Chemistry and Biochemistry at UCLA as an assistant professor in 1997, was promoted to associate professor in 2003, full professor in 2006, and received a joint appointment in the Department of Materials Science and Engineering in 2010. She is one of the founding members of the California NanoSystems Institute. Her research focuses on controlling nanometer scale architecture in solution processed nanostructured and nanoperiodic materials to generate unique optical, electronic, magnetic, structural, and electrochemical properties. While her group works in many areas, one key emphasis is on energy related issues–specifically nanomaterials for solar energy harvesting and electrochemical energy storage. She also leads a high school nanoscience program aimed at bringing related nano-concepts to high school students in the greater Los Angeles area. Tolbert is the recipient of a number of awards, including the Office of Naval Research Young Investigator Award, a National Science Foundation Faculty Early Career Development Award, a Beckman Young Investigator Award, a Sloan Foundation Research Fellowship, the UCLA Glen T. Seaborg Award, the UCLA McCoy Award, and a GRC/FCRP/NRI Inventor Recognition Award.

BRIAN D. WIRTH
Nuclear Engineering Department
University of Tennessee

Brian Wirth is Professor and Governor’s Chair of Computational Nuclear Engineering in the Department of Nuclear Engineering at the University of Tennessee, Knoxville, which he joined in July 2010. Wirth received a B.S. in nuclear engineering from the Georgia Institute of Technology in 1992 and a Ph.D. in mechanical engineering from the University of California, Santa Barbara in 1998, where he was a Department of Energy Nuclear Engineering Graduate Fellow. Wirth spent four years in the High Performance Computational Materials Science group at Lawrence Livermore National Laboratory, where he lead efforts to investigate the microstructural stability of structural materials in nuclear environments. In 2002, he joined the faculty at the University of California, Berkeley as an assistant professor of nuclear engineering and was promoted to associate professor in 2006. He has received a number of awards, including the 2007 Fusion Power Associates David J. Rose Excellence in Fusion Engineering Award and the 2003 Presidential Early Career Award for Scientists and Engineers (PECASE).

Professor Wirth’s research investigates the performance of nuclear fuels and structural materials in nuclear environments. This research will improve predictions about the longevity of nuclear reactor components and ultimately lead to the development of high-performance, radiation resistant materials for advanced nuclear fission and fusion energy power plants. The research approach involves an integrated and multi-disciplinary combination of computational multiscale materials modeling with experimental processing and characterization of materials structure and properties from the nanometer to continuum length scales to elucidate the dynamics of materials behavior. The research is funded by the U.S. Department of Energy, Office of Nuclear Energy, Office of Basic Energy Sciences and the Office of Fusion Energy Sciences; the National Science Foundation; and the Department of Homeland Security. Wirth has published more than 100 peer-reviewed papers in world known journals and conference proceedings, as well as organized/co-organized many national and international conferences.

LANS Science and Technology Committee Representatives

JOHN BERCAW

Centennial Professor of Chemistry
California Institute of Technology

John Bercaw received his B.S. degree from North Carolina State University in 1967, his Ph.D. from the University of Michigan in 1971, and undertook postdoctoral University of Chicago for one year. He joined the faculty at the California Institute of Technology as an Arthur Amos Noyes Research Fellow in 1972, and in 1974 he joined the professorial ranks, becoming professor of chemistry in 1979. From 1985 to 1990 he was the Shell Distinguished Professor of Chemistry, and in 1993 he was named Centennial Professor of Chemistry. Bercaw has been a Seaborg Scholar at Los Alamos National Laboratory (2004), the Robert Burns Woodward Visiting Professor at Harvard University (1999), Visiting Miller Professor at University of California, Berkeley (1990), and in 1989-90 a Royal Society of Chemistry Guest Research Fellow at Oxford University. Bercaw consulted with Exxon for more than 20 years and is now a consultant with Dow. He has served on numerous panels for the Department of Energy and the National Research Council, has been a member of the University of California, Office of the President's Panel on Science and Technology for Lawrence Livermore, Lawrence Berkeley and Los Alamos National Laboratories since 1999, and is a member of the LANS Science and Technology Committee. His research interests are in synthetic, structural and mechanistic organotransition metal chemistry. Recent studies include metallocene catalysts for Ziegler-Natta polymerization of olefins and investigations of hydrocarbon hydroxylation with transition metal complexes.

RACHEL A. SEGALMAN

Associate Professor
University of California, Berkeley

Rachel Segalman received her B.S. in chemical engineering with highest honors from the University of Texas at Austin. She then performed her doctoral work in chemical engineering (polymer physics) at the University of California, Santa Barbara. Following her Ph.D., Segalman was a postdoctoral fellow at the Universite Louis Pasteur in Strasbourg France. She then joined the faculty of UC Berkeley in the spring of 2004 as the Charles Wilke Assistant Professor of Chemical Engineering and was promoted to Associate Professor in 2009. Segalman's research focuses on the controlled self-assembly of polymer materials. In particular, her group has performed significant work in controlling the structure and thermodynamics of functional polymers including semiconducting and bioinspired polymers. She also has interest in designing polymeric and hybrid materials for energy applications involving thermoelectrics, photovoltaics, and solar fuels. She is an Alfred P. Sloan Fellow, a Camille Dreyfus Teacher Scholar, and has received the Presidential Early Career Award for Science and Engineering (PECASE), MDV Innovators Award, TR35: Technology Review's Top Innovators Under 35, Hellman Family Young Faculty Award, 3M Untenured Faculty Award, NSF CAREER Award, Intel Young Faculty Seed Award, and Chateaubriand Postdoctoral Fellowship. She is an associate editor for the *Annual Reviews of Chemical Engineering* and is on the editorial board of *Macromolecules.* Her website address is: http://www.cchem.berkeley.edu/rasgrp/

Materials Capability Review Theme Leaders and Presenters

RODNEY L. BORUP
Sensors and Electrochemical Devices (MPA-11)
Team Leader

Rod Borup is the program manager for the fuel cell program and is a team leader in the fuel cell program at Los Alamos National Laboratory. He received his B.S.E. in chemical engineering from the University of Iowa in 1988, and his Ph.D. from the University of Washington in 1993. He has worked on fuel cell technology since 1994, working in the areas of hydrogen production and PEM fuel cell stack components. He has been awarded 13 U.S. patents, authored ~~ 80 papers related to fuel cell technology and presented more than 100 oral papers at international and national meetings, with more than 1,300 citations. His current main research area is related to PEM fuel cell durability. He has been awarded the 2005 DOE Hydrogen Program R&D Award for his team's work in fuel cell durability and was the Principal Investigator for the 2004 Fuel Cell Seminar (San Antonio, TX USA) Best Poster Award.

WENDY R. CIESLAK
Materials Science & Technology (MST-DO)
Division Leader

Wendy R. Cieslak is the leader of the Materials Science & Technology (MST) Division at Los Alamos National Laboratory. MST Division, an organization of ~300 people and $90M, performs materials research and engineering for diverse national security challenges from nuclear weapons to energy security. The Division has specialized capabilities in processing of actinides, primarily plutonium and uranium, as well as precision machining for target fabrication, and a full spectrum of materials analysis and microscopy. Prior to joining Los Alamos in January 2009, Cieslak was deputy to the vice president for Science, Technology and Engineering (ST&E) at Sandia National Laboratories, where she led the Laboratory Directed R&D program, the University Partnerships program, and operations of the ST&E Strategic Management Unit. Her technical staff career started at Sandia in 1983 with basic and applied corrosion research of metals in liquid and atmospheric environments. She spent about a decade stewarding power sources from early R&D through to prototype production, specifically lithium/ thionyl-chloride battery development and lithium-ion battery research programs. Cieslak earned her Ph.D. and B.S. in materials engineering from Rensselaer Polytechnic Institute. A graduate Hertz Fellow, she was inducted to the Hertz Foundation board of directors in October 2008. She is a Fellow of ASM, International.

VICTOR I. KLIMOV
Physical Chemistry and Applied Spectroscopy (C-PCS)
Los Alamos National Laboratory Fellow

Victor Klimov is a Fellow of Los Alamos National Laboratory and the Director of the Center for Advanced Solar Photophysics of the U.S. Department of Energy. He received his M.S. (1978), Ph.D. (1981), and D.Sc. (1993) degrees from Moscow State University. He is a Fellow of the American Physical Society, a Fellow of the Optical Society of America, and a former Fellow of the Alexander von Humboldt Foundation. His research interests include optical spectroscopy of semiconductor and metal nanostructures, carrier relaxation processes, strongly confined multiexcitons, energy and charge transfer, photovoltaics, femtosecond and nonlinear optical spectroscopies, magnetic-field spectroscopy.

JENNIFER S. MARTINEZ
Center for Integrated Nanotechnologies (MPA-CINT)
Technical Staff Member

Jennifer Martinez is a technical staff member in the Center for Integrated Nanotechnologies at Los Alamos National Laboratory. She received a B.S. in chemistry from the University of Utah and a Ph.D. in bioinorganic chemistry (2002) from the University of California, Santa Barbara. Martinez received a Director's Funded Postdoctoral Fellowship (2002-2004) at Los Alamos to study biosensor development from discovery of recognition ligands by phage display to translational production of a deployable sensor (*J. Materials Chem*. **15** (2005); *Nuc. Acids. Res.* **34** *(2006); J. Immunol. Methods* **321** (2007); *Langmuir* **24** (2007)). Throughout her work she has studied the interaction between inorganic and biological systems in particular the colligative properties of iron-peptide amphiphiles (*Langmuir* **21** (2005); *P.N.A.S* **100** (2003); *J. Am. Chem. Soc.* **124** (2002); *Science* **287** (2000)); chemical synthesis using enzymatic catalysts (*J. Am. Chem. Soc.* **123** (2001)); natural product structure determination (*Limnol. & Oceanog.* **46** (2001)). Together with colleagues, Martinez has begun to utilize phage display (*Nuc. Acids. Res.* **34** (2006)) and biological and polyelectrolyte templating to create fluorescent metal nanoclusters (*J. Phys. Chem. C.* **111** (2007); *Nano Letters* **10(8)**, 3106-3110 *(*2010); *Chem. Commun.* **46**, 3280-3282 (2010); *Chem. Comm.*, DOI: 10.1039/C0CC03711G (2010); ," *J. P. C. C.* **114(38),** 15879-15882 (2010)). Professional honors and service include the LANL LDRD-ER review subcommittees "Synthetic and Analytical Chemistry" (2004); "Materials and Methods" (2005), "BBB" (2006; chair: 2009, 2010, 2011), and "Chemistry and Chemical Sciences" (2008); ad hoc service on NIH and USDA study sections; lead a highly successful and recognized LDRD-DR on gold and silver nanocluster development with more than eight publications and two patents at mid-term review; Presidential Early Career Award for Science and Engineering (2008); Outstanding Mentoring Award (2007), Directors Postdoctoral Fellow, and graduate of the Leadership Development Initiative, LANL; B.R. Baker Award in Chemistry, California SeaGrant Traineeship and Graduate Opportunity Fellowship, UCSB.

DUNCAN W. McBRANCH
Science, Technology & Engineering (PADSTE)
Deputy Principal Associate Director

Duncan McBranch is the deputy principal associate director for Science Technology and Engineering. He has been an R&D leader in materials chemistry and nanotechnology, an entrepreneur in biotechnology, and a business development leader in growing new programs at Los Alamos with industry.

McBranch was division leader for Technology Transfer at Los Alamos (2005–2007), where he was responsible for collaborations with industry, for the commercialization of inventions, and for the spin-out of regional technology businesses. Previously, McBranch led teams solving complex research, technology, and business problems for over 15 years. This includes founding a biotechnology company (QTL Biosystems) to develop rapid assays for the life sciences markets and handheld detection solutions for environmental pathogen detection. He began his technical career at Los Alamos as a Director's Postdoctoral Fellow. After become a staff member, he led a research team investigating polymeric materials for nanotechnology with applications in optics and electronics.

McBranch has published more than 70 articles in technical journals in materials, chemistry, and biotechnology, and he is an inventor with patents across a broad range of applications. He earned undergraduate degrees in physics and mathematics (Whitman College) and a Ph.D. in materials physics (University of California, Santa Barbara).

MICHAEL NASTASI

Center for Integrated Nanotechnologies (MPA-CINT)
Los Alamos National Laboratory Fellow

Michael Nastasi is the director of the Energy Frontier Research Center on Materials at Irradiation and Mechanical Extreme at Los Alamos National Laboratory. He received his B.S. (1981), M.S. (1983) and Ph.D. (1986) degrees from the Materials Science and Engineering Department at Cornell University and has been at Los Alamos since 1985. His research interests include ion-solid interactions, irradiation induced phase transformations, ion irradiation and plasma modification of materials, ion beam analysis of materials, synthesis and properties of high strength nanolayered composites, and surface mechanical properties. Nastasi's work in these areas has resulted in several awards including the 1995 Los Alamos National Laboratory Fellows Prize for his extensive research in ion-solid interactions and the R&D 100 Award in 1997 for "Plasma Source Ion Implantation for Enhancing Materials Surfaces." He was appointed a Los Alamos National Laboratory Fellow in the summer of 2000, was made Fellow of the American Physical Society in 2006, and appointed Fellow of MRS in 2011. He has served as an adjunct professor at Arizona State University, the University of Maryland, and the University of Colorado, and has been a visiting professor at the University of Helsinki in Finland. He has co-authored more than 470 refereed publications, which have been cited more than 6,000 times, authored the books *Ion-Solid Interactions: Fundamentals and Applications*, published by Cambridge University Press in 1996 and *Ion Implantation and Synthesis of Materials*, published by Springer-Verlag, Berlin in 2006, and edited several volumes including the MRS bestseller, the *Handbook of Modern Ion Beam Materials Analysis*.

ANDREW T. NELSON

Materials Science and Technology (MST-7)
Research Scientist

Andrew T. Nelson is a research scientist in the Materials Science and Technology (MST) Division at Los Alamos National Laboratory. He received his B.S. in engineering mechanics followed by his M.S. and Ph.D. in nuclear engineering from the University of Wisconsin-Madison. His research interests are focused on the study of thermophysical properties of both metals and ceramics at high temperatures, with an emphasis on materials for nuclear applications and development of advanced experimental techniques. Nelson is also active in the characterization of materials' performance in spallation environments as well as the design of materials systems for spallation target applications. Currently, Nelson is the ceramic fuels irradiation testing lead within the DOE-NE Fuel Cycle Research and Development program.

JOHN L. SARRAO

Science Program Office-Office of Science (SPO-SC)
Program Director

John Sarrao is the program director for Los Alamos National Laboratory's Office of Science Programs, a $100M/y portfolio, and for MaRIE (Matter-Radiation Interactions in Extremes), LANL's signature facility concept that will provide transformational materials solutions for national security challenges. Since 2002, John has held leadership positions of increasing responsibility within LANL's materials community. He has also served on a number of U.S. Department of Energy Basic Energy Sciences Advisory Committee (BESAC) Subcommittees, helping to set strategic directions for materials research. Sarrao received his Ph.D. in physics from the University of California, Los Angeles in 1993 based on thesis work performed at LANL. He returned to LANL as a technical staff member in 1997 following postdoctoral research with Zachary Fisk at the University of California, San Diego and the National High Magnetic Field Laboratory in Tallahassee, Florida. His primary research interest is in the synthesis and characterization of correlated electron systems, especially actinide materials. He is the coauthor of more than 520 publications, including 56 papers in *Physical Review Letters*, *Nature*, and *Science*. These publications have been cited more than 10,000 times (h=50). He was the 2004 winner of the LANL Fellows Prize for Research, in part for his discovery of the first plutonium superconductor and is a Fellow of the American Association for the Advancement of Science (AAAS), the American Physical Society (APS), and Los Alamos National Laboratory.

KURT SCHOENBERG

Los Alamos Neutron Science Center (LANSCE)
User Facility Director
Experimental Physical Sciences (EPS)
Deputy Associate Director

Kurt Schoenberg is presently the user facility director of the Los Alamos Neutron Science Center (LANSCE) and deputy associate director of Experimental Physical Sciences at Los Alamos National Laboratory. In these capacities, he oversees the operations of basic and applied research performed at the LANSCE facility. This includes neutron scattering research at the Lujan Neutron Scattering Center, nuclear science and technology at the WNR facility, and oversight of the research program at the proton radiography facility. He received his BS in engineering physics with high honors from the University of Illinois in 1972. In 1979, he was awarded a PhD in physics from the University of California, Berkeley and joined the Los Alamos National Laboratory's research staff. Schoenberg's research expertise and accomplishments, as documented by more than 90 publications, include the experimental and theoretical investigation of magnetically confined plasmas for controlled thermonuclear fusion, inertial fusion, intense particle accelerators, plasma accelerators, plasma-based space propulsion, missile interceptor systems, and high-energy-density-physics.

SUSAN SEESTROM

Experimental Physical Sciences (ADEPS)
Associate Director

Susan Seestrom brings to this position a combination of strong science credentials and management skills developed during her 20-year tenure at LANL. In her recent role as associate director of Weapons Physics, Seestrom led six LANL divisions that executed program work in experimental, simulation, and weapons physics assessment. She directed the major line organization responsible for carrying out research and development for the weapons program and technical work in support of stockpile certification and assessment. She oversaw the operation of complex facilities, including the Dual-Axis Radiographic Hydrodynamic Test Facility, the Los Alamos Neutron Science Center, and the U1a laboratory at the Nevada Test Site.

Previously, Seestrom led the Physics Division at LANL for three years after serving as a deputy group leader in the Neutron Science and Technology group. Seestrom's personal research efforts focused on nuclear structure with medium energy probes and fundamental physics with neutrons. Together with Laboratory scientist Tom Bowles, Seestrom led the effort to develop an ultracold neutron source that culminated in demonstration of the world's most intense source of such neutrons. She received a Distinguished Performance Award for this work in 2001.

Seestrom holds Ph.D. and B.S. degrees in physics from the University of Minnesota. She has published 135 papers and has had 1,663 career citations. She is a Fellow of the American Physical Society.

JACK SHLACHTER

Theoretical Division (T-DO)
Deputy Division Leader

Jack Shlachter graduated from the California Institute of Technology in 1975 with a B.S. and received his Ph.D. in physics from University of California, San Diego in 1982. His research focused initially on experimental plasma physics, exploring dense z-pinch configurations for magnetic fusion applications. For several years he served as a lead experimenter on the Pegasus Pulsed Power facility, studying material strength issues in dynamic implosions, and he was the chief scientist for the successor

Atlas high current facility at Los Alamos. Additional research in fusion has included the examination of magnetized target fusion geometries. He has held a variety of line management positions in Physics Division and led the Division from 2004-2008. Following a leave of absence to the Comprehensive Test Ban Treaty Organization in Vienna during 2008, he began an involvement with the planning efforts for the MaRIE (Matter-Radiation Interactions in Extremes) signature facility, and he currently works on the Fission Fusion materials Facility as a member of the core MaRIE team. For the past year he has been the deputy division leader of Theoretical Division.

CHRIS STANEK

Structure/Property Relations (MST-8)
Staff member

Chris Stanek received his B.S. in materials science and engineering at Cornell University where he was a McMullen Scholar and his Ph.D. in materials from Imperial College London (under the supervision of Prof. Robin Grimes). His research interests focus on the interaction between multidimensional defects in ceramics, primarily via atomistic simulation techniques. Stanek has a particular interest in materials for nuclear energy, including transmutation fuels, crystalline waste forms and scintillator radiation detectors. Currently, Stanek is the materials performance optimization focus area lead of the recently awarded Energy Innovation Hub for the Advanced Modeling and Simulation of Nuclear Reactors. Stanek has published more than 40 papers related to defect behavior in ceramics.

ANTOINETTE (TONI) TAYLOR

Materials Physics and Applications (MPA-DO)
Division Leader

Antoinette Taylor is the leader of the Materials Physics and Applications Division at LANL. Prior to this position she was director of the Center for Integrated Nanotechnologies, a joint Sandia/LANL Nanoscience Research Center funded through BES. Her research interests include the investigation of ultrafast dynamical nanoscale processes in materials and the development of novel optics-based measurement techniques for the understanding of new phenomena. She has published more than 250 papers in these areas, written 2 book chapters and edited 4 books. She is a former director-at-large of the Optical Society of America, topical editor *of Journal of the Optical Society B: Optical Physics*, member-at large, Division of Laser Science of the American Physical Society and member of the Solid State Science Committee, Board of Physics and Astronomy, the National Academies and she chaired the National Academies' Committee on Nanophotonics Applicability and Accessibility. She is chair and OSA's representative to the Joint Council on Quantum Electronics. She is a LANL Laboratory Fellow and a Fellow of the American Physical Society, the Optical Society of America and American Association for the Advancement of Science. In 2003, Taylor won the inaugural Los Alamos Fellow's Prize for Outstanding Leadership in Science and Engineering.

DAVID F. TETER

Materials Science & Technology (MST-DO)
Deputy Division Leader

David Teter is the Materials Science and Technology (MST) deputy division leader. MST Division provides world-leading, innovative, and agile materials science and technology solutions for national security missions.

After finishing his doctoral thesis at the University of Illinois in 1996, he began a postdoctoral appointment at Los Alamos National Laboratory researching hydrogen storage and solid-state phase transformations in Pd-based alloys. In 1997 he was converted to a full-time technical staff member in the Metallurgy Technology group (MST-6). As a staff member, he expanded his research interests into the areas of alloy design, hydrogen storage, hydrogen-induced phase changes, solid-state phase transformations and aging phenomena of weapons materials.

In 2002 Teter became a weapons project leader for metals issues. In this role, he was responsible for the technical direction and planning of the program leading to several key decisions regarding material re-use and remanufacturing. This project led to his next role in 2006 as the project leader for the Enhanced Surveillance CSA/Case effort. The main focus of this program is to understand and quantitatively predict lifetimes of materials, components and assemblies. This project combines fundamental scientific research of aging mechanisms and kinetics with engineering assessments of performance.

Concurrently, Teter was the team leader for the Alloy Design and Development Team from 2003 to 2005, which primarily investigated processing-structure-property relationships of materials as they relate to engineering and physics requirements. From 2005-2010, he was the deputy group leader of MST-6 where he led a diverse technical organization, which uses materials technology to support national security.

Teter also serves as the chair for the M4 (Making, Measuring and Modeling Materials) pillar of the Matter-Radiation Interaction in Extremes (MaRIE) experimental facility, which is vital to many national security challenges and is a critical component of the LANL Materials Strategy. The M4 facility will provide the experimental, modeling and research tools to accelerate materials discovery, control synthesis and design of materials, and address the decadal materials challenges of the future. Teter is also a member of the Materials Science and Engineering Alumni Board at the University of Illinois.

TERRY WALLACE

Science, Technology & Engineering (PADSTE)
Principal Associate Director

Terry Wallace is the principal associate director of Science, Technology, and Engineering, which is responsible for all basic science programs at Los Alamos, and coordinates the activities of the four science and engineering directorates. During the period of 2005 to June 2006, Wallace was the associate director of the Strategic Research Directorate, which encompassed LANL's science program offices and the five line divisions that implemented those programs and supported LANL's nuclear weapons, threat reduction, and energy security missions. He was also responsible for LANL's non-National Nuclear Security Administration Department of Energy programs, including basic science, energy technology, and environmental technology. Before becoming the associate director for Strategic Research, Wallace was the division leader of the Earth and Environmental Sciences Division.

Raised in Los Alamos, Wallace returned in 2003 after 20 years as a professor of geosciences and an associate in the applied mathematics program at the University of Arizona. In addition to teaching, he carried out research on global threat reduction, nonproliferation verification, and computational geophysics. During his academic career, he worked with LANL on nuclear test monitoring and threat reduction, in particular on interpreting the indications of nuclear testing by a foreign government. He has an international reputation in geosciences as applied to national security issues. Wallace holds Ph.D. and M.S. degrees in geophysics (California Institute of Technology) and B.S. degrees in geophysics and mathematics (New Mexico Institute of Mining and Technology). He is the author or coauthor of more than 80 peer-reviewed publications on seismology and tectonics, including ground-based nuclear explosion monitoring and forensic seismology. He also wrote a widely used textbook on seismology. Wallace is a Fellow of the American Geophysical Union (AGU), and in 1992 he received the AGU's Macelwane Medal. Wallace has served as president of the Seismological Society of America, chairman of the Incorporated Institutions for Research in Seismology, and authored the position paper for the American Geophysical Union on the verifiability of a comprehensive test ban treaty. He has testified before Congress on the comprehensive test ban and participated in numerous National Academy panels, including ones on research in support of comprehensive test ban monitoring. From 2000-2006, Wallace was the chair on the National Research Council's Committee on Seismology and Geodynamics.

YONGQIANG WANG

Materials Science in Radiation and Dynamics Extremes Group (MST-8)
Technical Staff Member and Director of Ion Beam Materials Laboratory

Yongqiang Wang received his Ph.D. in nuclear physics and technology from Lanzhou University (China) in 1992. He then spent the next 10 years working at Missouri State University (Postdoc Fellow/Research Assistant Professor), University of Louisiana – Lafayette (Research Scientist/Research Assistant Professor), and University of Minnesota – Twin Cities (Senior Research Associate/Ion Beam Analysis Facility Manager). After joining LANL in 2003, he has been managing the Ion Beam Materials Laboratory (IBML), a LANL resource devoted to materials research through the use of energetic ion beams. His primary research interests include materials characterization using ion beam analysis techniques, materials modification and synthesis using ion implantation technology, and radiation damage effects in materials induced by energetic ion beam bombardments. He has also operated and maintained four different Tandem accelerators and four different ion implanters in five accelerator facilities in China and the USA. He has authored or coauthored more than 120 peer-reviewed publications including 3 book chapters, 1 U.S. patent, and 1 *Handbook of Modern Ion Beam Materials Analysis*- Second Edition (2009). In 2006, he initiated a new topic area on radiation damage effects for the Biennial International Conference Series on Application of Accelerators in Research and Industry (CAARI), and has served as its topic editor for *CAARI-2006, CAARI-2008* and *CAARI-2010*. He is a co-chair of Biennial International Conference on Ion Beam Analysis (IBA-21) to be held in Seattle in 2013.

DAVID WATKINS

Materials Physics and Applications (MPA-DO)
Deputy Division Leader

David Watkins is deputy division leader for the Materials Physics and Applications Division. Watkins got his B.S. in physics at New Mexico Tech, and his M.S. and Ph.D. in physics from the University of Washington. He joined the staff at Los Alamos in 1979 to work on nonlinear optics for the laser fusion and isotope separation programs. He later worked on the Strategic Defense Initiative, exploring issues of high-power laser beam propagation through the atmosphere. As part of this effort, Watkins spent a year at the Royal Signals and Radar Establishment in Great Britain as part of a technical exchange on the Strategic Defense Initiative. Following this appointment, he returned to Los Alamos and became involved in the management of the Electronic and Electrochemical Materials group (now MPA-11). To facilitate the energy research carried out in this group, Watkins developed and managed more than 20 cooperative research agreements with industry and played a key role in increasing the size of the fuel cell research program at Los Alamos. He next took on the challenge of program manager for Laboratory Directed Research and Development at Los Alamos. In this position, he worked to foster a strong program in basic research at LANL that enhances the scientific vitality of the Laboratory in critical, mission-relevant, areas of research and development. He returned to MPA Division with the goal of contributing to a leading research organization engaged in the critical issue of energy security.